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BENCH-SCALE ASSESSMENT OF MEMBRANE PRE-TREATMENT AND
SEASONAL FOULING VARIATIONS

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ÉCOLE POLYTECHNIQUE DE MONTRÉAL

Ce mémoire intitulé:

BENCH-SCALE ASSESSMENT OF MEMBRANE
PRE-TREATMENT AND SEASONAL FOULING VARIATIONS

présenté par : ESQUIVEL PÉREZ, ANA GABRIELA

en vue de l'obtention du diplôme de : Maîtrise ès sciences appliquées

a été dûment accepté par le jury d'examen constitué de :

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À Antoine

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RÉSUMÉ

La filtration membranaire est une technologie versatile qui est considérée comme une solution potentielle à plusieurs défis auxquelles on fait face aujourd'hui dans le traitement des eaux. La nanofiltration est très intéressante pour les petits systèmes qui recherchent des alternatives pour l'enlèvement de la matière organique naturelle (MON) autre que le traitement conventionnel. Cependant, le colmatage limite la productivité des membranes et reste une problématique importante pour l'opération des systèmes membranaires. Beaucoup de recherches ont été faites sur les causes possibles du colmatage et l'identification du meilleur prétraitement pour le prévenir mais les recherches publiées à ce jour, se sont surtout concentrées sur des prétraitements avancés qui sont prohibitifs pour les petits systèmes, et les études de colmatage utilisent typiquement l'eau synthétique ou des eaux fractionnées pour étudier les possibles agents colmatants.

Dans le but d'investiguer le colmatage par les eaux naturelles, deux eaux de surface (le Lac Barron et la Rivière des Prairies) ont été évaluées en termes des variations de pouvoir colmatant et de qualité d'eau entre février et octobre 2007. Le pouvoir colmatant a été quantifié avec le MFI-UF (modified fouling index – ultrafiltration) et des mesures de turbidité, des comptes des particules, de carbone organique totale (COT), d'absorbance UV₂₅₄, de couleur, d'absorbance UV spécifique (SUVA), des concentrations de Ca, Mg, Fe et de conductivité ont été faites en parallèle. L'objectif était d'investiguer l'impact des variations saisonnières de qualité d'eau sur le potentiel colmatant des eaux par l'exploration des relations apparentes entre le MFI-UF et les paramètres de qualité complémentaires. Des analyses statistiques faites par analyse en composantes principales (ACP) et par régression « partial least squares » (PLS) ont montré que l'influence de la qualité d'eau sur le potentiel colmatant dépend de la source de l'eau et que les paramètres traditionnels ne sont donc pas de bons indicateurs généraux. Cependant, quand on parle de sources spécifiques certains paramètres contribuent de façon importante au

développement des modèles prédictifs : la turbidité, les comptes des particules et les concentrations de fer pour la Rivière des Prairies et la turbidité, le COT et la concentration de fer pour le Lac Barron ont été identifiés comme des variables importantes pour le développement des modèles prédictifs avec des coefficients de détermination (R^2) élevés.

En plus du suivi saisonnier, la performance des filtres à lavage automatique comme moyen de réduction du pouvoir colmatant a été jaugé. Des échantillons de tamis fins (25 μm and 10 μm) et des filtres en textile (2 μm et 3 μm) ont été utilisés afin de simuler la performance que cette technologie pourrait avoir dans une installation à grande échelle. La réduction du potentiel de colmatage a été quantifiée par le MFI-UF et la réduction des particules par des comptes de particules. L'hypothèse était que l'étape de préfiltration enlèverait les particules plus grandes que les porosités annoncées des préfiltres et qu'une plus grande réduction des particules se traduirait par une plus grande réduction du MFI-UF. Les comptes des particules ont montré que l'enlèvement des particules plus grandes que le degré de filtration annoncé n'a pas été absolu et les mesures de MFI-UF faites en parallèle ont démontré que l'enlèvement des particules observé (0.5 log pour le filtre de 2 μm) n'a pas causé une diminution appréciable du pouvoir colmatant. Il a été donc conclu que les particules de tailles supérieures à 2 μm ne jouent pas de rôle important dans le colmatage des membranes. Des expériences ont été réalisées pour déterminer le rôle que la formation d'un gâteau sur la surface d'une membrane joue sur l'enlèvement des particules. Il était soupçonné que l'enlèvement des particules allait s'améliorer pendant la filtration mais les comptes des particules ont montré que ça n'était pas le cas pour les particules plus grandes que 2 μm . Il y a eu une légère réduction de turbidité et il est donc probable qu'il y ait eu une petite amélioration dans l'enlèvement des particules submicroniques. La formation d'un gâteau n'a pas causé une réduction appréciable dans le pouvoir colmatant.

Les résultats de cette étude ont des implications pratiques pour l'implémentation des systèmes membranaires. Les corrélations spécifiques trouvées pour les différentes sources et leurs relations aux événements climatiques peuvent être appliquées à la planification des essais pilotes pour s'assurer que les essais sont faits pendant les conditions de colmatage les plus critiques. Les résultats suggèrent également qu'il est inutile d'essayer d'utiliser les paramètres de qualité traditionnels pour prédire le colmatage sans avoir auparavant établi un profil de colmatage pour la source en question. L'évaluation du prétraitement a montré que même si les filtres à lavage automatique ont été capables d'enlever les particules plus grandes que 2 μm pour les eaux étudiées ici, ils n'ont pas été capables de réduire le colmatage des coupons de membrane, même avec l'enlèvement des solides le plus fin.

ABSTRACT

Membrane filtration is a highly versatile technology which has recently gained much popularity as a potential solution to many of the challenges currently being faced in water treatment. Nanofiltration seems particularly promising for small systems looking for alternatives to conventional treatment for the removal of NOM. Fouling however, limits membrane productivity and is a major concern for the operation of membrane systems. Much research has been done on trying to better understand the causes of fouling and on identifying the best pre-treatments to prevent it. The work published however has focused on advanced pre-treatments that are prohibitive to small systems and fouling studies typically have use synthetic or fractioned waters to investigate possible foulants.

In order to investigate fouling by natural waters, two surface waters (Barron Lake and the Des Prairies River) were evaluate in terms of fouling potential and water quality variations from February to October 2007. Fouling potential was quantified using the modified fouling index- ultrafiltration (MFI-UF) and additional quality parameters measured included turbidity, particle counts, TOC, UV254, color, SUVA, conductivity and Ca, Mg and Fe concentrations. The objective was to investigate the impact of seasonal water quality variations on the fouling potential of the waters by exploring any apparent relationships between the MFI-UF and the complementary quality parameters. Statistical analysis of the results using principal component analyses (PCA) and partial least squares regressions (PLS) showed that the influence of water quality on fouling potential was source dependent, and that traditional parameters did not make good general fouling indicators for this reason. When referring to specific water sources however certain parameters contributed heavily to the development of a successful explanatory models; turbidity, particle counts and iron concentrations for the Des Prairies River and turbidity, TOC and iron concentrations for Barron Lake were found to be

important predictors in the development of prediction models with high coefficients of determination (R^2).

In addition to the seasonal fouling assessment, the performance of automatic backwash filters as a fouling potential pre-treatment was evaluated at a bench-scale. Samples of fine screens (25 μm and 10 μm) and cassette filters (2 μm and 3 μm) were used to simulate the performance this technology would have in full scale installations. Fouling potential reduction was again quantified by means of the MFI-UFs and particle removal was evaluated by means of particle counts. The hypotheses were that the pre-filtration step would remove particles larger than the announced pre-filter porosities and that a greater degree of particle removal would bring with it a greater degree of MFI-UF reduction. Particle counts showed that the removal of particle larger than announced filtration degrees was not absolute, and MFI-UF measurements done in parallel demonstrated that the removal of particles observed (about 0.5 log for the 2 μm filter) did not lead to an appreciable reduction in fouling potential. It was therefore concluded that particles larger than 2 μm do not play an important role in fouling of the flat sheet membrane themselves. Experiments were also conducted to investigate the role that the formation of a cake layer on the surface of a 2 μm cassette filter would have on particle removal. While it was thought that the removal of particles would improve as the filtration run progressed and the cake layer built it, particle counts showed that this was not the case for particles larger than 2 μm . There was a slight decrease in turbidity, and it is therefore probably that there was a small improvement in the removal of sub-micron particles. The formation of the cake layer did not cause an appreciable reduction in fouling potential either.

The results of this study have practical implication on the implementation of membrane systems. The source specific fouling correlations found and their relation to weather events could be applied to the planning of membrane piloting schedules at specific locations to test units under the most challenging conditions. Results also suggest that

attempting to use traditional water quality parameters to predict fouling without first establishing a fouling profile of the water in question would be fruitless. The pre-treatment evaluation showed that while automatic backwash filtration technologies may be somewhat successful at removing particles from feed waters like those studied here, they did not have reduce the fouling of flat sheet membrane, even with the finest solids removal options.

CONDENSÉ EN FRANÇAIS

La demande croissante pour une eau de haute qualité combinée avec la nature limitée des sources disponibles a causé une demande pour le développement de technologies avancées capables d'enlever des polluants émergents telles que les toxines algales, les sous produits de désinfection et les produits pharmaceutiques. La filtration membranaire est une de ces technologies et elle est une solution prometteuse qui permet de faire face aux défis de qualité d'eau auxquelles on fait face aujourd'hui. Au Canada, il y a plusieurs petits systèmes qui traitent l'eau des lacs fortement chargée en matière organique naturelle. Ces petits systèmes font face aux problématiques mentionnées ci-dessus et en plus ont souvent un accès limité aux ressources telles que du personnel qualifié. La nanofiltration est une option intéressante pour ces petits systèmes puisqu'elle peut être utilisée pour faire face à des défis variés tout en restant une technologie compacte, automatisée et relativement indépendante des additifs chimiques.

Le défi le plus important pour l'opération réussie de la filtration membranaire est le colmatage. Le colmatage est une problématique complexe qui n'est encore ni bien comprise ni bien quantifiée par les paramètres traditionnels de qualité d'eau. Les indices de colmatage ont été créés pour aider à résoudre ce problème, mais la compréhension des facteurs qui jouent un rôle dans le colmatage reste incomplète.

Le prétraitement peut être utilisé pour limiter le colmatage mais les options de prétraitement extensif sont typiquement prohibitives pour les petits systèmes. La sélection du prétraitement en amont de la filtration membranaire doit être basée sur les mêmes critères qui rendent les membranes une solution intéressante. Le besoin de construire des infrastructures importantes, de doser des produits chimiques en continu ainsi que des coûts de capital et d'opération élevés sont considérés comme les contraintes les plus importantes pour l'implémentation des traitements par des petits systèmes. Les

manufacturiers de membranes demandent typiquement une certaine protection contre les particules pour l'opération des modules membranaires (i.e. l'installation de filtres en cartouche ou leur équivalent en amont des membranes), alors la protection contre les particules est un critère nécessaire pour un prétraitement membranaire.

Un classement des différents traitements disponibles basé sur les critères mentionnés ci-dessus est présenté dans la revue de littérature. Ce classement a démontré que l'option la plus intéressante à étudier plus en détails est la filtration à lavage automatique.

Objectifs

L'objectif général du projet est de mieux comprendre le colmatage par des eaux naturelles de surface et sa prévention par le prétraitement physique afin de faciliter l'utilisation des membranes spiralées de haute pression dans des petites communautés. Le projet est divisé en deux sections, la première section étudie les variations saisonnières du colmatage et la deuxième section étudie l'impact des prétraitements sur la réduction du pouvoir colmatant.

Matériel et Méthodes

Deux eaux de surface ont été étudiées de février à octobre 2007, la Rivière des Prairies (Laval) et le Lac Barron (Lachute). Seulement l'eau de la Rivière des Prairies, qui est la plus chargée des deux en particules, a été utilisée dans la section d'évaluation de prétraitement.

Le pouvoir colmatant des eaux a été mesuré avec un indice de colmatage, le MFI-UF (modified fouling index, ultrafiltration). Cet indice est basé sur la théorie de filtration sur gâteau qui dit que la résistance hydraulique est causée par la formation d'un gâteau de

particules qui sont retenues à la surface de la membrane. Le MFI-UF est une fonction de la concentration et de la nature des particules de l'influent et il correspond à la pente de la portion linéaire de la courbe de t/V vs V . Cet indice est mesuré avec une membrane de UF de 10kDa fait en polycrylonitrile à une pression constante de 207 kPa (30 psi) et à température pièce. Un nouveau coupon de membrane a été utilisé pour chaque expérience et tous les coupons utilisés ont été soumis à un préconditionnement pour limiter la variabilité des perméabilités initiales entre les différents coupons. Ce conditionnement a consisté en une trempage dans l'eau ultrapure pour au moins 12 heures suivi par une compaction à une pression de 310 kPa (45 psi) avant de mesurer la perméabilité initiale du coupon à 207 kPa (30 psi) avec l'eau ultrapure.

Plusieurs paramètres de qualité d'eau ont été mesurés en plus du MFI-UF pour tous les échantillons provenant des deux sources dans le cadre de la première section de l'étude. Ces mesures incluent : la turbidité, des comptes des particules de tailles supérieures à 2.25 μm , les concentrations de Fe, Mg et Ca, la conductivité, le carbone organique total (COT), la couleur, l'absorbance UV à 254 nm et l'absorbance UV spécifique (SUVA). Ces paramètres ont été analysés en conjonction avec les mesures de pouvoir colmatant afin de mieux comprendre le colmatage.

L'analyse des données du suivi saisonnier a été faite avec deux méthodes statistiques : l'analyse des composantes principales (ACP) et la régression « partial least squares ». Le ACP est une méthode exploratoire multivariable qui est utile pour la classification des variables et des observations et qui est donc utile pour l'interprétation des données. La régression PLS est une méthode de régression qui peut être utilisée quand les méthodes traditionnelles de régression ne sont pas applicables comme quand il y a peu d'observations et plusieurs variables colinéaires, comme dans ce cas-ci.

Les pré-filtrations effectuées pour la deuxième section du projet ont servi à déterminer la performance des filtres à lavage automatique en termes de la réduction de pouvoir colmatant. Elles ont été faites avec des échantillons du matériel filtrant utilisé dans les filtres à échelle industrielle fabriqués par la compagnie Amid. Deux sortes des filtres ont été utilisées, des tamis en acier inoxydable de 10 et 25 μm et des filtres en textile de 2 et 3 μm . Deux aspects de la filtration ont été étudiés, l'effet de la porosité du préfiltre sur le colmatage et les comptes des particules et le rôle que la formation d'un gâteau sur le filtre joue sur le colmatage et les comptes des particules. Le deuxième aspect a été étudié en échantillonnant le filtrat d'un seul filtre en textile de 2 μm à différents moments du cycle de filtration.

Résultats Préliminaires

Comme mentionné auparavant, les coupons de membrane utilisés ont été prétraités afin de limiter la variabilité des perméabilités initiales. Les coupons ont été rejetés si leur perméabilité initiale variait par plus que 20% de la valeur annoncée par le manufacturier $1.94 \times 10^{-7} \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1} / \text{kPa}$ (70 l/h/bar), ou s'ils n'arrivaient pas à atteindre une perméabilité stable durant la dernière heure et demie de filtration.

Les résultats de perméabilité présentés ici n'incluent pas les coupons qui ont été rejetés parce qu'ils n'ont pas respecté les critères énumérés ci-dessus. Trente-deux coupons ont été utilisés pour le suivi saisonnier et neuf coupons ont été utilisés pour l'évaluation du prétraitement, tous les coupons ont été coupés du même m^2 de membrane. La perméabilité moyenne de ces coupons a été de $1.74 \times 10^{-7} \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1} / \text{kPa}$ ($62.71 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) et l'écart type a été de $1.55 \times 10^{-8} \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1} / \text{kPa}$ ($5.59 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$). La perméabilité moyenne a été 10% plus basse que celle annoncée par le manufacturier.

Avant de décider d'utiliser des échantillons de préfiltres utilisés dans l'industrie, l'utilisation des filtres de laboratoire a été considérée pour déterminer quelle taille de particules joue le rôle le plus important dans le colmatage. Des filtres en polypropylène faits par la compagnie Millipore de différentes porosités (0.6 μm , 1.2 μm , 2.5 μm , 5 μm , 10 μm , 30 μm) ont été utilisés pour préfiltrer des échantillons d'eau brute de la Rivière des Prairies. Au moment des filtrations il a été remarqué que les turbidités des filtrats étaient étranges. Le filtrat produit avec le filtre de 1.2 μm avait une turbidité supérieure au filtrat produit avec le filtre de 30 μm . En faisant des comptes des particules, il a été trouvé que les filtres qui performaient le mieux en termes d'enlèvement des particules étaient les filtres de 2.5 μm (1.64 logs), de 5 μm (1.17 μm logs), de 30 μm (0.88 logs) et de 10 μm (0.81 logs). Les filtres qui avaient le pire enlèvement des particules étaient les filtres de 0.6 μm (0.45 logs) et 1.2 μm (0.30 logs).

Suite à une répétition des filtrations pour vérifier que la source d'erreur n'était pas humaine, des essais ont été faits pour vérifier si les inconsistances étaient causées par un relâchement des particules par les filtres ou par des particules déjà présentes dans le compteur des particules. Le protocole de nettoyage pour le compteur des particules a été revisité et il a été vérifié que les filtres ne relâchaient pas des particules. Puisqu'aucune des deux possibles sources d'erreur n'a expliqué les résultats de façon acceptable, il a été conclu que c'était probablement un défaut de manufacture qui avait causé la mauvaise performance des filtres et c'est pourquoi il a été décidé de ne pas les utiliser.

Résultats Principaux

Suivi Saisonnier

Comme on pouvait s'y attendre, le profil de qualité du Lac Barron a été très différent de celui de la Rivière des Prairies. Les différences seront discutées plus en détails un peu plus loin.

Il est important de noter que les particules de taille de $2.5 - 3 \mu\text{m}$ ont toujours représenté la fraction la plus importante pour les comptes des particules pour le Lac Barron ainsi que pour la Rivière des Prairies. Le pourcentage moyen des particules qui avaient cette taille a été $63\% \pm 6\%$ et $68\% \pm 4\%$ pour chaque source respectivement. Les particules de tailles supérieures à $10 \mu\text{m}$ ont été systématiquement la fraction la moins importante avec des pourcentages moyens de $5\% \pm 4\%$ et $3\% \pm 1$ pour le Lac Barron et pour la Rivière des Prairies. Les particules de tailles supérieures à $30 \mu\text{m}$ ont représenté en moyenne moins que 0.5% de la concentration totale des particules pour les deux sources.

Une analyse ACP a été effectuée avec les paramètres de qualité traditionnels pour les deux sites, pour mieux jauger s'il serait plus approprié d'analyser les résultats des deux sites ensemble ou individuellement. Les résultats de l'ACP ont montré que les échantillons des deux sources varient de façon prédominante selon deux facteurs (variables composites représentatives) différentes. La régression PLS qui a été faite par la suite a montré que la source de l'échantillon était la variable la plus importante pour la création d'un modèle prédictif par cette méthode. Donc les analyses statistiques faites en suite ont été faites de façon individuelle pour chaque source d'eau.

Les ACP faites pour chaque source ont élucidés les corrélations entre plusieurs des paramètres mesurés. Le COT, l' UV_{254} , la couleur et la turbidité étaient corrélés pour les

échantillons pris au Lac Barron, tous les paramètres avaient des points au début de juin et à la fin d'octobre. La turbidité, conductivité, concentrations de Ca, Mg, et Fe ainsi que les comptes des particules avaient des intercorrélations pour les échantillons de la Rivière des Prairies et ils avaient tous des points à la mi-avril pendant la période de fonte des neiges.

En général les paramètres associés à la matière particulaire et colloïdale étaient significativement plus élevés pour les échantillons provenant de la rivière que du lac. Les paramètres associés au contenu ionique (Fe, Mg, Ca) avaient des valeurs légèrement plus élevées pour les échantillons de l'eau de rivière et les paramètres reliés à la matière organique avaient des valeurs comparables pour les deux sites. Étonnamment, les plages des valeurs du MFI-UF étaient très semblables pour les deux sources malgré les différences importantes dans les signatures des types d'eau.

Une régression « PLS » a été faite pour chaque type d'eau afin d'investiguer toute relation apparente entre le MFI - UF et les paramètres de qualité d'eau traditionnels qui ont été mesurés. Les modèles produits ont été capables de prédire les valeurs de MFI - UF avec des modèles qui avaient des coefficients de détermination (R^2) proche de 0.90 pour les deux sites d'échantillonnage. Pour le modèle bâti avec les observations faites au Lac Barron, les variables qui avaient l'influence la plus importante sur la variabilité du MFI-UF étaient la turbidité, le COT et la concentration de Fe. La couleur et l'absorbance UV_{254} ont joué un rôle qui était légèrement moins important.

Pour le modèle bâti avec des observations faites à la Rivière des Prairies, les variables les plus importantes pour la prédiction du MFI-UF étaient la turbidité, les comptes des particules et la concentration de Fe. La conductivité et les concentrations de Mg ont contribué au modèle aussi mais dans une plus faible mesure.

Prétraitement

Effet de la porosité du préfiltre sur le MFI-UF et les comptes des particules.

Comme mentionné auparavant quatre préfiltres (2 μm , 3 μm , 10 μm et 25 μm) ont été utilisés pour déterminer quel rôle la porosité du préfiltre joue sur la réduction du MFI-UF et l'enlèvement des particules. Les filtres de 2, 3 et 10 μm ont été capables d'enlever respectivement 74%, 82% et 66% des particules avec des diamètres supérieurs aux porosités annoncées. Les résultats pour le filtre de 25 μm n'ont pas été présentés car les concentrations des particules de diamètre supérieure à 25 μm dans les échantillons d'eau brute utilisés pour cette série d'analyses ont été négligeables (< 50 particules /ml).

Les préfiltres ont réduit la turbidité de façon marginale, même pour la porosité la plus serrée (2 μm). La turbidité de l'échantillon d'eau brute était 3.12 UTN tandis que les filtrats des filtres de 2 μm , 3 μm , 10 μm et 25 μm avaient des turbidités de 2.36 UTN, 2.55 UTN, 2.66 UTN et 2.67 UTN respectivement. Les filtrations n'ont pas eu d'effet significatif sur les valeurs de MFI-UF. Les variations entre les valeurs d'eau filtrée et d'eau brute ont toujours été de $\pm 10\%$ et selon les chercheurs qui ont développé l'indice, une variation de moins que 10 % peut être considérée comme marginale.

Effet de la formation d'un gâteau sur le MFI-UF et les comptes des particules

L'enlèvement des particules de diamètres supérieures à 2.25 μm a diminué légèrement avec la formation d'un gâteau en surface du filtre en textile de 2 μm . Le pourcentage d'enlèvement est passé de 89% à 87% et finalement à 83% pendant la durée de la filtration. L'échantillon d'eau brute utilisé pour cette série d'expériences avait une turbidité de 9.6 UTN tandis que les échantillons qui ont été collectés aux volumes

spécifiques de $0.2 \text{ m}^3/\text{m}^2$, $0.7 \text{ m}^3/\text{m}^2$ et $1.2 \text{ m}^3/\text{m}^2$ avaient des turbidités de 4.70 UTN, 3.83 UTN et 3.34 UTN respectivement. Même si l'enlèvement des grandes particules ($> 2.25 \text{ }\mu\text{m}$) avait diminué avec le temps, l'amélioration de la turbidité indique un enlèvement amélioré des petites particules par le gâteau. Pourtant, même avec le gâteau qui a été formé sur le cassette pendant la filtration, les MFI-UF des échantillons pris à des volumes spécifiques de 0.2, 0.7 et $1.2 \text{ m}^3/\text{m}^2$ n'ont pas varié de façon importante par rapport aux valeurs de l'échantillon de l'eau brute ($33\,000 \text{ s/L}^2 \pm 12\%$). Même si ces variations sont légèrement plus élevées que le seuil de 10 %, elles ne peuvent pas être classifiées comme étant des variations majeures.

Discussion

Résultats Préliminaires

À cause de la petite taille des coupons utilisés pour la mesure du MFI-UF (13.4 cm^2), la nature hétérogène des feuilles de membrane peut causer des variations dans les données de perméabilité initiale. Même avec conditionnement, plusieurs coupons ont été rejetés à cause des perméabilités excessivement basses. Aussi, la perméabilité moyenne des coupons qui ont été retenus a été plus basse que la perméabilité annoncée par le fabricant. Il est possible que ce phénomène soit dû au fait que les spécifications des fabricants s'appliquent typiquement aux modules membranaires et non à des petites surfaces de membrane. La petite quantité d'information qui est donnée par les fabricants est un défi pour le développement des approches standardisées et des travaux futurs devraient proposer des guides pour amener un certain niveau d'uniformité aux essais de laboratoire.

Les résultats des essais préliminaires faits avec des membranes de laboratoire en polypropylène ont montré qu'il n'est pas toujours possible de supposer avec certitude que

les filtres de laboratoire vont performer de la façon qui est annoncée. Des vérifications doivent être faites quand cela est possible si des expériences vont être faites avec des filtres de laboratoire.

Suivi Saisonnier et analyse du pouvoir colmatant

Les résultats des analyses statistiques faites sur les variations de qualité d'eau et sur le potentiel colmatant suggèrent que les paramètres de qualité d'eau traditionnels ne peuvent pas être utilisés de façon générale pour déterminer ni la présence ni la concentration de polluants. Cependant, les modèles qui ont été développés pour une source d'eau en particulier ont réussi à expliquer la variabilité du potentiel colmatant en fonction des paramètres mesurés. Ceci implique que l'influence de la qualité d'eau sur le MFI-UF et donc sur le potentiel colmatant est dépendante de la source d'eau.

Dans le cas du Lac Barron, il y avait une forte corrélation entre le COT et le potentiel colmatant, ceci suggère que les agents colmatants pour cette source (possiblement des fractions spécifiques de la MON) variaient en proportion avec le COT. De la même façon la turbidité était fortement corrélée avec le potentiel colmatant du Lac Barron et le potentiel colmatant de la Rivière des Prairies, suggérant que la fraction spécifique de la turbidité qui jouait un rôle important dans le colmatage variait en proportion avec la turbidité totale dans les deux cas.

La caractérisation du Lac Barron et de la Rivière des Prairies pendant une période de huit mois a facilité le développement des profils de qualité pour les deux sources fournissant donc un contexte pour mieux interpréter les variations de potentiel colmatant. La caractérisation peut servir à donner de l'information sur l'importance que des événements météorologiques peuvent avoir sur le potentiel colmatant (i.e. fonte des neiges, fleur d'eau). Les travaux futurs devraient se concentrer sur l'identification et le développement

de méthodes de quantification qui ciblent les possibles agents colmatants (matière colloïdale, les protéines et polysaccharides, etc.) dans les influents. En parallèle, une procédure standard pour créer des profils des sources utilisant des mesures subrogées (turbidité et COT au Lac Barron) devrait être créée pour suivre l'évolution du pouvoir colmatant de façon simple à travers l'année.

Évaluation du prétraitement

Les comptes des particules ont montré que les préfiltres n'ont pas complètement enlevé les particules des tailles supérieures aux porosités annoncées. Cependant, certains des filtres ont systématiquement montré un enlèvement non négligeable, le filtre de 2 μm , par exemple, a enlevé 74% ou plus de particules des tailles supérieures à 2.25 μm . Cet enlèvement n'a pas eu d'effet important sur la réduction du MFI-UF. Il semble alors que l'enlèvement des particules plus grandes que 2 μm joue un rôle négligeable sur la réduction du potentiel colmatant.

Il était attendu que le gâteau qui se forme sur la surface d'un filtre allait avoir un effet sur l'enlèvement des particules par le filtre. Pourtant, la formation d'un gâteau sur un filtre en textile de 2 μm à débit constant n'a pas amélioré l'enlèvement des particules de diamètres supérieures à 2.25 μm . Il y a même eu une légère augmentation dans les comptes des particules vers la fin de la filtration, probablement à cause des forces de cisaillement qui se sont développées. La diminution de turbidité observée suggère que le gâteau a amélioré l'enlèvement des particules plus petites que la limite de détection du compteur des particules (2.25 μm). L'enlèvement des petites particules n'a pas été suffisant pour altérer de façon significative le potentiel colmatant des échantillons de filtrat.

Il devrait être noté que même si l'enlèvement des particules n'a pas donné des avantages en termes de réduction du colmatage des membranes, il est probable que la protection contre les grandes particules limite l'obstruction des modules spiralés. Il semble peu probable par contre qu'une préfiltration grossière serait utile pour cette protection car les concentrations des particules plus grandes que 30 μm étaient typiquement très basse comparé avec les concentrations des autres tailles des particules pour les deux sources étudiées. Puisque le colmatage ne semble pas être affecté par les grandes particules il est probable que des colloïdes sont responsables pour le colmatage et que le prétraitement pour membranes devrait être capable d'enlever les colloïdes pour réduire le pouvoir colmatant de manière effective. Les travaux futurs devraient se concentrer sur l'investigation des méthodes de traitement capables d'enlever la matière submicronique sans trop augmenter les coûts d'opération.

Conclusions

L'objectif principal du projet a été atteint. L'investigation des impacts des variations saisonnières de qualité sur le pouvoir de colmatage a fait ressortir le fait que les paramètres de qualité traditionnels sont inadéquats pour décrire ou quantifier les agents colmatant de façon générale mais que ces mêmes paramètres peuvent être utiles pour suivre le potentiel colmatant quand les profils des eaux spécifiques sont connues. L'évaluation de la réduction du pouvoir colmatant par des filtres à lavage automatique a été aussi une réussite. Il a été déterminé que la taille des particules enlevées par cette technologie (2 μm et plus) ne joue pas de rôle dans le colmatage et donc que le colmatage des membranes ne peut pas être réduit avec ce traitement.

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CHAPITRE 1 : INTRODUCTION

1.1 Context for research

Recently there has been a growing awareness of the limited availability of water resources and the possible health risks associated with compounds that may be found in potable water. In order to respond to these emerging issues, water treatment technologies have evolved and changed. Membrane technologies are at the forefront of this movement and they are being hailed as a versatile solution for many of the current concerns.

While in Canada, water shortage is generally not a severe issue as it is elsewhere on the globe, there are still water quality issues that need to be addressed. The successful delivery of safe drinking water in smaller communities is one of the most challenging issues faced in Canada. Smaller water producers often have to cope with limited financial, technical and human resources that complicate the operation of chemical dependant conventional treatment. Nanofiltration (NF) is an alternative to conventional treatment for the removal of NOM (natural organic matter), a widely recognized DBP precursor and source of color.

Even though NF appears promising, there are still many challenges to operating these membrane systems, the most important of which is fouling. Fouling is a complex issue and is presently neither well understood nor well quantified through traditional water quality parameters. Fouling indexes have been created to address this shortcoming but there is still an incomplete understanding of the factors which play a role in fouling.

Pre-treatment can be used to mitigate fouling but extensive pre-treatment options are usually prohibitive for smaller systems. The selection of the pre-treatment upstream of

nanofiltration needs to be based on the same criteria that make NF an attractive solution. NF membrane manufacturers typically demand a certain degree of protection from particles for the successful operation of their membrane modules (i.e. the installation of cartridge filters or equivalent upstream of membranes). The need to build important infrastructure, to dose chemical additives regularly as well as large capital and operating costs are considered as being important constraints to consider for the selection of pre-treatment in small NF systems.

A ranking of different pre-treatment schemes is presented further on in the literature review (table 2.1). Based on the criteria described above, automatic backwash filters were found to be the most interesting option and were therefore selected as the pre-treatment technology to be further studied.

1.2 Project description

This study is a part of a larger inter-university study funded by the Canadian Water Network which aims at advancing membrane processes for Canadian drinking water treatment facilities. The study presented here is composed of two sections; the first looks at the possible causes behind fouling using two natural water sources while the second evaluates the performance of the automatic backwash filter technology with the water source that was found to be most heavily charged with particles.

1.3 Project objective

The general objective of this project is to gain a better understanding of fouling by natural surface waters and its prevention through physical pre-treatment in order to facilitate the use of spiral wound high pressure membrane filtration systems in small communities. The project is divided into two sections which respectively study the issue of seasonal variations in fouling potential and the impact of pre-treatment to reduce the later.

1.4 Report structure

The report is comprised of 6 chapters. This first chapter is the introduction, chapter 2 presents a focused literature review on fouling of NF membranes and pre-treatment options, chapter 3 briefly critiques previous work done and presents the objectives and hypothesis supporting this research, chapter 4 is an article that will be submitted to the Journal of Membrane Science, chapter 5 presents additional results that were not discussed in chapter 4, chapter 6 includes the general discussion of the results, recommendations for future work and a brief conclusions.

CHAPITRE 2 : LITERATURE REVIEW

As water resources diminish in quantity and quality and regulations become more and more stringent, alternative treatment technologies have become crucial in dealing with emerging water quality issues. Membrane technology has recently garnered much attention for being an extremely promising technology capable of addressing many of the water quality and quantity issues that we face today. Its small footprint, high level of automation and its relative independence from chemical additives (Mallevialle et al., 1996) distinguish it from other treatment technologies. Stricter disinfection by-products (DBP), cyanotoxins and micro-pollutants regulations, as well as the need to exploit less than pristine water sources due to water scarcity, have made membrane filtration a rising star in the world of water treatment.

2.1 Generalities

A membrane is a semi-permeable barrier that is used to selectively limit the transport of undesirable matter (Mallevialle et al., 1996). In water treatment, the undesirable materials which need to be removed from the finished water ranges from dissolved contaminants such as salts to particulate matter such as microbial contaminants.

While membrane filtration was initially used for seawater desalination it has since diversified and now has widespread applications in wastewater, groundwater and surface water treatment. Different families of membrane products have been developed to better satisfy the different applications; spiral wound high pressure reverse osmosis (RO) and nanofiltration (NF) for the removal of dissolved and small colloidal matter and hollow

fiber low pressure microfiltration (MF) and ultrafiltration (UF) for the removal of larger particulate matter, microorganisms and coagulated matter. It should be noted that the distinction between the different membrane types is not absolute and there is no consensus as to what the delimitations between the categories are.

Membrane families have different separation mechanisms. Low pressure membranes (MF/UF) have larger pores and therefore steric exclusion dominates, while exclusion by affinity dominates for dense RO membranes. NF membranes, which lie somewhere between RO and UF, display both steric and affinity exclusion mechanisms (Mallevialle et al., 1996), the extent to which a mechanism dominates is a function of the tightness of the membrane being used. With looser NF membranes steric exclusion dominates while with tighter NF membranes diffusion exclusion dominates.

Nanofiltration can be used to address a variety of issues affecting drinking water producers at lower pressures and therefore lower pumping costs than RO. Removal of disinfection by-product precursors is particularly problematic for plants treating surface waters. In recent years, new membrane packages geared towards smaller systems have made this technology more accessible to smaller communities. Membrane filtration is fast becoming a viable solution for these communities to meet water quality regulations that previously required chemical treatment (Anderson et Sakaji, 2007). NF is particularly interesting for small treatment systems whose source water is colored lake water rich in NOM (natural organic matter) who are having difficulty meeting disinfection by-product regulations (Kouadio et T  rault, 2003).

It is widely recognized that NF can successfully reject dissolved organic matter, a disinfection by-product precursor of organo-chlorinated compounds. Agbekodo et al., (1996), Orecki et al. (2004) and Thanuttamavong et al. (2002) all reported high TOC and/or DOC rejection by NF membranes. High rejections of TTHM (total

trihalomethanes) and HAA9 (nine haloacetic acid species) precursors by several commercially available NF membranes have also been reported (Chellam, 2000).

NF is also an interesting option for cyanotoxin removal (Ellis, 2007). Recently, the issue of cyanobacteria blooms has received increased attention worldwide. Promising results on the rejection of microcystine – LR and anatoxin by NF membranes have been published (Gijssbertsen-Abrahamse et al., 2006)

Nanofiltration is also being looked at as a potential treatment for the removal of EDC/PPCP (endocrine disrupting compounds and personal care products). (Yoon et al., 2007) observed rejection of various pharmaceutical compounds to varying degrees by NF membranes.

2.2 Fouling

While membrane filtration looks full of promise there are still hurdles that have to be overcome. The most important challenge in the implementation of membrane technologies remains membrane fouling. In order to advance membrane technologies and exploit them to their fullest potential, membrane fouling must be addressed and managed (Escobar et al., 2005).

Fouling is the undesirable accumulation of material (foulants) in the pore structure, on the surface of the membrane and in the feed spacer. It reduces membrane permeability by constricting the pores and/or by forming a layer whose resistance may be higher than that of the membrane itself. Reduced permeability leads to decreases in permeate flux or increases in transmembrane pressure and thus increases pumping costs, membrane

cleaning frequency and overall operation and maintenance costs (Mallevialle et al., 1996; Boerlage et al., 1997; Her et al., 2007). Membrane fouling is a complex issue that is function of the feedwater composition, physical-chemical properties of the foulants, membrane characteristics (material, roughness, charge and hydrophobicity) and the interactions between all these factors (Escobar et al., 2005).

Different membrane families are fouled by different mechanisms. Low pressure membranes have larger pores than high pressure membranes and therefore fouling of UF and MF membranes is dominated by pore adsorption and plugging which are mechanisms that are likely to be less important for NF membranes (Hong et Elimelech, 1997).

The agents found in feedwater that are responsible for fouling and the mechanisms through which they reduce flux are typically classified into four categories: particulate fouling, organic fouling, scaling and biofouling (Boerlage et al., 1997). In practice however the different types of fouling tend to occur in conjunction influencing one another in ways that are not presently well understood (Spettmann et al., 2007) for example the role that organic matter may play in the cohesion of colloids in the formation of a gel layer (Mallevialle et al., 1996).

It is also evident that the distinction between particulate/colloidal, organic, biological and precipitative fouling of membranes is not sharp and that there exists a certain degree of overlap (Yiantsios et al., 2005). For example, there is some confusion surrounding biofouling due to distinction between biotic fouling (caused by an active biofilm) and abiotic fouling (caused by natural organic matter (NOM) of microbial origin) (Escobar et al., 2005). NOM of microbial origin is considered more as a part of organic fouling by some (Her et al., 2007), and more as biofouling by others (Speth et al., 2000). In addition to this when discussing organic fouling, the nuance between colloidal and truly dissolved organic matter is not made. This is because the distinction between particulate and

dissolved matter has traditionally been made based on retention by a 2 μm filter, and because the definition of dissolved organic matter is based on filtration on a 0.45 μm filter. This simplified definition does not take colloids into account (Amy et al., 2001). And it may be difficult to distinguish between fouling by adsorption and cake formation with complex mixtures like NOM (Hong et Elimelech, 1997)

Much work has been done trying to better understand and manage fouling, however many of the studies which have been done approach the problem using the typical fouling classification system and therefore often use laboratory water spiked with one or two model foulants making it difficult to take into account all the interactions which make fouling so complicated. Many of these studies which aim at better understanding the causes of fouling of NF membranes have focused on trying to identify which fraction or fractions of dissolved NOM are responsible for fouling of NF membranes. NOM is a highly heterogeneous mixture of particulate and soluble components of different molecular weights and chemical compositions which can be divided into hydrophobic (humic), hydrophilic and transphilic substances (Zularisam et al., 2006). Thus far, the results of these different studies have been conflictive and tend to isolate the effects of dissolved NOM from particulate/larger colloidal matter. (Nilson et DiGiano, 1996) concluded that the humic fraction of waters was responsible for fouling while other studies report that the hydrophilic components such as proteins and polysaccharides, possibly of microbial origin, caused the most significant fouling (Jarusutthirak et al., 2002; Park et al., 2006).

Studies looking at particulate/colloidal fouling are often guilty of the same exclusivity as the studies on organic fouling. Studies conducted using laboratory inorganic colloid solutions and NF membranes (Boussu et al., 2007) and even laboratory water spiked with inorganic colloids and NOM (Kim et al., 2007), are not necessarily representative of the colloidal/particulate fouling observed with natural waters. (Kweon et Lawler, 2004)

found that between natural and laboratory waters with comparable DOC's, the laboratory water (which had a significantly higher turbidity) caused less severe fouling of tight UF membranes and that synthetic waters with no DOC and very high turbidities caused almost no flux decline.

Precipitative fouling and biotic biofouling have not been discussed in detail here since the study at hand focuses on the measurement of short term fouling by two natural surface waters from Québec, Canada. Precipitative fouling or scaling is the formation of scales on the membrane when salts in the raw water are concentrated on the feed side beyond their solubilities. This process is on a very slow time scale when compared to particulate/colloidal fouling (Brauns et al., 2002). Biotic biofouling is a biofilm phenomenon (Flemming et al., 1997) and biofilm formation is critical in systems with feedwaters above 25 °C (Al-Ahmad et al., 2000) which is rarely the case in Canada.

2.3 Fouling indexes

Since the causes and mechanisms of fouling are not well understood, it is difficult to estimate the fouling potential of natural waters using traditional water quality parameters. Measurements such as turbidity and particle counts have been used as fouling indicators but are not considered as reliable predictors (Yiantsios et al., 2005; Boerlage et al., 2000; Roorda et van der Graaf, 2005). Other measurements of water quality such as DOC, UV₂₅₄ and SUVA have also been used but no relationship was found between membrane fouling and these parameters (Howe et Clark, 2002a; Howe et Clark, 2006; Heijman et al., 2005). A distinction will be made here between indicators which are parameters (such as particle counts) that are used to indirectly measure fouling potential and indexes which quantify fouling through some direct measure of flux decline.

Fouling indexes have been developed to quantify fouling potential and fill this gap. Most are bench-scale membrane tests that provide a simple and relatively inexpensive method to conduct an initial screening before moving on to more costly and time consuming pilot testing (DiGiano et al., 2000). Care should be used when utilizing these indexes as they describe an intrinsic characteristic of a water sample and are not design parameters. The conditions under which the fouling indexes are determined are often quite different from the operating conditions of a full scale system and they typically do not capture membrane - solute interactions which are specific to each membrane (Khirani et al., 2006; (Roorda et van der Graaf, 2005). They remain however useful tools.

The two most commonly used fouling indexes are the SDI (silt density index) and the MFI (modified fouling index) (Montgomery Watson Harza (MWH), 2005) which will be described in the following paragraphs.

2.3.1 Silt Density Index (SDI)

The SDI is a longstanding standardized index which was developed to quantify the plugging potential a water would have on a RO membrane. It is calculated based on the two time intervals which are required to filter fixed volumes of sample through a 0.45 μm membrane at a constant transmembrane pressure of 207 kPa bar (30 psi) (ASTM International, 2002). The SDI is an empirical test with no theoretical background and since it is measured with a 0.45 μm membrane it excludes the effect of smaller colloids (Boerlage et al., 1998). This index therefore provides very limited information on fouling. Poor correlations between fouling of NF and RO membranes and SDI measurement have been reported in the literature (Koyuncu et al., 2006); Chellam et al., 1997; Yiantisios et al., 2005).

2.3.2 The Modified Fouling Index (MFI)

The MFI is a fouling index developed by (Schipper et Verdouw, 1980) which is based on cake filtration, one of the four theoretical fouling mechanisms established by (Hermia, 1982). In this fouling mechanism, the retention of particles on the surface of the membrane increases the hydraulic resistance encountered during filtration. These accumulated particles and colloids form a cake layer which adds its resistance, to that of the membranes in a resistance in series model. It should be noted that this model was developed for constant pressure filtration and that the increase in hydraulic resistance is proportional to the cumulated volume of filtered water V .

The interest in using a fouling index which is based on cake filtration is that fouling of NF and RO membranes is presumed to occur mainly through a cake filtration mechanism due to the extremely small size of their pores (Boerlage et al., 2002). And while the MFI is a dead end filtration test, it has been reported that flux decline in a cross-flow regime is similar to that measured in dead end filtration (Koyuncu et al., 2006).

The MFI is a function of the nature and concentration of the particles in the sample. A typical filtration curve of a t/V vs V graph is shown below, the MFI is the slope of the linear portion of the graph which corresponds to the cake filtration region.

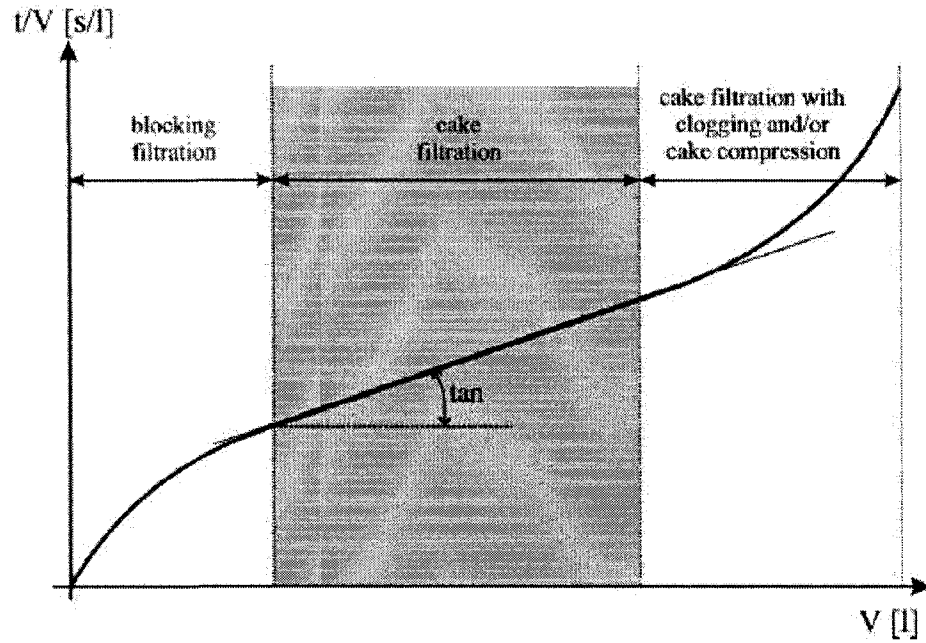


Figure 2-1: Typical filtration curve (t/V vs V)

(Boerlage et al., 2003b)

The MFI is derived as follows:

$$J = \frac{\Delta P}{\eta(R_{tot})} = \frac{\Delta P}{\eta(R_m + R_c)} \quad (\text{Equation 2-1})$$

$$J = \frac{dV}{dt} \times \frac{1}{A_m} \quad (\text{Equation 2-2})$$

$$\frac{dV}{dt} \times \frac{1}{A_m} = \frac{\Delta P}{\eta(R_m + R_c)} \quad (\text{Equation 2-3})$$

Where:

$$R_c = \frac{V}{A_m} \times \alpha C_b = \frac{V}{A_m} \times I \quad (\text{Equation 2-4})$$

and

$$\alpha = \frac{180(1 - \epsilon)}{\rho_p d_p^2 \epsilon^3} \quad (\text{Equation 2-5})$$

$$\frac{dV}{dt} = \frac{\Delta P A_m}{\eta(R_m + \frac{V}{A_m} I)} \quad (\text{Equation 2-6})$$

Integrating equation 2-6 over constant pressure.

$$\frac{t}{V} = \frac{\eta R_m}{\Delta P A_m} + \left(\frac{\eta I}{2 \Delta P A_m^2} \right) V = \frac{\eta R_m}{\Delta P A_m} + (MFI) V \quad (\text{Equation 2-7})$$

Where:

J is flux ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$)

t is filtration time (s)

V is the permeate produced (m^3)

ΔP is the transmembrane pressure (N m^{-2})

η is the dynamic viscosity of water (N s m^{-2})

A_m is the membrane area (m^2)

R_m is the membrane resistance (m^{-1})

R_c is the cake layer resistance (m^{-1})

I is the propensity of particles to form a hydraulic resistant layer (m^{-2})

α is the specific cake resistance (m kg^{-1})

C_b is the particle concentration (kg m^{-3})

MFI-UF is the modified fouling index (s/L^2)

The reference conditions for the test measurements are a constant pressure of 207 kPa (30 psi) and a temperature of 20°C (Schipper et Verdouw, 1980).

Since the MFI and MFI-UF (discussed further below) are measured with membranes that do not reject salts, the osmotic pressure is not taken into account by this index. The osmotic pressure of macromolecules and colloidal species is usually quite small (Mallevialle et al., 1996).

2.3.3 The Modified Fouling Index – Ultrafiltration (MFI-UF)

The MFI-UF follows the same principal as the MFI and in fact the only difference between the indexes lies in that a UF membrane is used to measure the MFI-UF instead of the 0.45 μm membrane. The use of a tight UF membrane incorporates the effects of colloids on fouling which would otherwise be missed with tests like the SDI and the traditional MFI. Work done by (Boerlage et al., 2002) determined that the membrane most suitable for MFI-UF measurement was a 13 kDa polyacrylonitrile (PAN) membrane since the MFI-UFs found using PAN membranes were stable over time.

2.3.4 The Unified Fouling Index

The unified fouling index (UMFI) was developed to quantify and assess the fouling of low pressure hollow fiber membranes. Like the fouling indexes mentioned above, it is based on the cake layer formation equation, the differences between the UMFI and the aforementioned indexes are that it is measured using a constant flowrate rather than a constant pressure and that it does not have a specified reference membrane. The fact that

this index is measured using the membrane of choice permits it to take into account the membrane – foulant specific relationships (Jacangelo, 2006; Huang, 2008).

2.3.5 Other fouling indexes

Several fouling indexes and fouling monitoring techniques have been proposed in the literature in addition to the ones detailed above, the MFS (membrane fouling simulator) (Vrouwenvelder et al., 2006), the SUR (specific ultrafiltration resistance) (Roorda et van der Graaf, 2005) and the MFI-NF (modified fouling index – nanofiltration) (Khirani et al., 2006) to name a few. Of all the indexes mentioned, the MFI-UF was chosen as the fouling index of most interest for the present study for multiple reasons. Firstly because its theoretical base, the cake filtration theory is the mechanism thought to dominate in the fouling of high pressure membranes. Secondly the suggested reference membrane takes the effects of colloids into account, and thirdly it has been used in other works looking at the effect of pretreatment (Boerlage et al., 2000; Tamas, 2004) and therefore it was interesting to use the same fouling index for the cross comparison between results to better understand the scale of the values found.

2.4 Membrane feed pretreatment

The research being done on membrane fouling focuses as much on the development of better tools to understand and quantify fouling as it does on its management. Fouling control is an important challenge in the operation of membrane systems. Different approaches can be taken to deal with this challenge including physical and/or chemical pretreatment of the membrane feedwater, membrane selection, membrane cleaning methods and frequencies, membrane module design and modification of operation parameters such as recovery rate, filtration rate and number of trains (Fane et al., 2000).

The focus of this literature review is on pretreatment, and the discussion of other management techniques will be limited as each technique could be the subject of its own literature review. Improved pre-treatment generally leads to better membrane performance, lower fouling and therefore lower overall operation and maintenance costs.

Prevention of colloidal and organic fouling is highly dependent on effective pretreatment. Selection of pretreatments upstream from NF membranes treating surface waters is particularly crucial for the success of the process since the characteristics of surface waters can vary significantly and have higher turbidities than ground waters in general (Kim et al., 2007).

The mitigation of fouling through improved pretreatment is not a new concept. In fact much work has been done in this area. However, the majority of the studies published have focused on extensive treatment methods that require developed infrastructure, trained personnel, chemical addition or other proposed methods that are very expensive, making the suggested solutions inaccessible to small communities. These options typically require on line chemical dosing (coagulation and/or ozonation) or additional low pressure membrane systems upstream of the high pressure membrane systems.

Loosely speaking any membrane systems with some sort of pretreatment is referred to as an integrated membrane system (IMS). In the drinking water community IMSs refer to RO/NF preceded by additional treatment processes such as coagulation- sedimentation- filtration (CSF), riverbank filtration, slow sand filtration (SSF), biological activated carbon (BAC), MF or UF and any combination of the above (Schippers et al., 2004).

Many studies have been conducted to compare the fouling potential reduction of these advanced pretreatments and there is no consensus as to which is the best one. (Glucina et al., 2000) found that both conventional and UF treatment produced good quality feed

water, (Chua et al., 2003) reported that low-pressure membrane filtration produced better quality feed water more consistently than traditional treatment and (Kim et al., 2007) found that in some cases MF outperformed traditional treatment while in others CSF outdid MF. Additional treatments reported in the literature with various degrees of success in the reduction of fouling of NF membranes include powdered activated carbon (PAC), ozonation (Lee et Lee, 2007), biological filtration (Mosqueda-Jimenez et Huck, 2006; Koyuncu et al., 2006), riverbank filtration (Nederlof et al., 2000) and UV treatment (Koyuncu et al., 2006). More advanced treatment trains have also been the subject of research; CSF/O₃/BAC/SSF was found to produce an effluent whose fouling potential decreased after each stage of pre-treatment, (MFI-UF's of 3600 s/L², 3200 s/L², 2500 s/L² and 400 s/L² respectively). The same study looked at CSF/UF (MFI-UF of 1700 s/L²) which produced feedwaters of lower fouling potential than CSF alone (MFI-UF of 8500 s/L²) (Boerlage et al., 2000).

Needless to say, such intensive pre-treatment is prohibitive and most probably unnecessary for small systems whose source water is of good quality. A ranking of pre-treatments options based on criteria that would make them accessible to small systems is presented in table 2-1. These criteria included of automation, independence from chemical additives and low costs. Automatic backwash filters came up at the top of the list due to the protection they offer from particles and the ease of operation they offer at a lower cost than low-pressure membranes (the runner up). While research that used coarse pre-filtration screens (200 µm and 50 µm) as pre-treatment for membrane modules reported insufficient protection from particles (Drage et al., 2001), other studies have reported successful operation of NF systems with automatic backwash filters (3 µm and 15-25µm respectively) as the only particle barrier (Nemirovsky, 2006; Ericsson et al., 1996). These studies however did not describe treatment performance in terms of fouling potential, but only in terms of turbidity and particle reductions, and at times not even that.

Typically, NF suppliers demand a 5 μm filtration upstream of their membranes as part of their processes guarantee (Nemirovsky, 2006). It is required as a safety measure so that upsets in pretreatment upstream do not clog membrane spiral wound modules. While (Koyuncu et al., 2006) reported that filtration through a 5 μm cartridge filter did not reduce fouling of NF or RO flatsheet membranes, the feed water used had already been extensively pre-treated (CSF/O³/GAC) and therefore it cannot be concluded that a 5 μm pre-filtration would not reduced the fouling potential of raw waters.

Table 2-1: Ranking of pre-treatment technologies

Technology	Advantages	Disadvantages	Rank
Automatic backwash filters	<ul style="list-style-type: none"> • Compact technology • Automated • Removes particles 	<ul style="list-style-type: none"> • Shorter lifespan (in the case of textile) • Fairly expensive in capital costs 	1
Low pressure membrane filtration (MF/UF)	<ul style="list-style-type: none"> • Compact technology • Automated • Removes particle and colloids 	<ul style="list-style-type: none"> • Expensive option • Fouling problems • Pumping costs 	2
Rapid sand filtration	<ul style="list-style-type: none"> • Removes particles • Automated 	<ul style="list-style-type: none"> • Requires important infrastructures • Possible damage to membranes due to sand • Better performance with coagulation • Often followed by cartridge filtration (see below) 	3
Cartridge filters	<ul style="list-style-type: none"> • Removes particles • Simple to install and operate 	<ul style="list-style-type: none"> • Expensive option • Disposable technology (permanent replacement purchase) • Not automated (labor intensive) 	4
Bag filters	<ul style="list-style-type: none"> • See cartridge filters • Less expensive than cartridge filtration 	<ul style="list-style-type: none"> • Not automated (labor intensive) 	5
Biological filtration	<ul style="list-style-type: none"> • Removes particles • Removes assimilable organic carbon 	<ul style="list-style-type: none"> • Better performing with ozone addition • Expensive option (GAC) • Requires important infrastructures (see sand filter) 	6
Slow sand filtration	<ul style="list-style-type: none"> • Removes particles • Simplicity of maintenance 	<ul style="list-style-type: none"> • Large footprint • Limited production (m^3/m^2) • Low efficiency in cold climates 	7
Coagulation/Flocculation/ Sedimentation	<ul style="list-style-type: none"> • Removes NOM and colloid removal 	<ul style="list-style-type: none"> • Needs on-line addition of chemical products • Needs important infrastructures • Needs qualified personnel 	8

CHAPITRE 3 : OBJECTIVES AND HYPOTHESES

3.1 Critique of previous work done

As underlined in the literature review, many of the published studies (Boussu et al., 2007; Lee et al., 2005; Nilson et DiGiano, 1996) focus on observing individual fouling mechanisms either by using synthetic waters or by fractioning natural waters into different components before studying each component separately. This does not provide a comprehensive picture of fouling since synthetic waters do not necessarily behave the same way natural waters do (Kweon et Lawler, 2004) and because there is often an overlap and a certain synergy between the fouling types (Spettmann et al., 2007).

Since the mechanisms and components behind fouling are not well understood at this point, traditional water quality parameters have not been found to well represent the fouling potential of water samples (Yiantsios et al., 2005; Howe et Clark, 2002a). Standardized fouling indexes have been developed to address this problem and allow for quantification of fouling. While many indexes have been proposed, the MFI-UF (modified fouling index – ultrafiltration) was found to be the most interesting as it has been shown to well represent cake filtration which is the fouling mechanisms thought to dominate in high pressure membrane filtration (Boerlage et al., 2002). It is also essentially the same as a commonly used index, the MFI, with the exception of using a tight UF membrane instead of a MF membrane.

The pre-treatment schemes that have most often been proposed in the literature as solutions for the control of membrane fouling in high pressure membrane systems (Kim et al., 2007; Lee et Lee, 2007; Boerlage et al., 2000) are methods that require either on

line chemical injection, heavy infrastructure or additional membrane systems upstream of the NF systems. These constraints are often prohibitive for small systems.

While there have been reports of successful operations of membrane systems with pre-treatment techniques which are simpler to operate (i.e. automatic backwash filters) (Nemirovsky, 2006; Ericsson et al., 1996), these works did not quantify the improvements in operation that were gained by introducing the pre-treatment stage. Quantifications of changes in fouling potential are particularly important for determining if this technology is effective at reducing fouling for the membranes downstream.

3.2 Objectives

3.2.1 Seasonal water quality and fouling potential analysis

The main objective of this sub-project is to investigate the impact of seasonal water quality variations on the fouling potential of two natural waters during an eight month period by means of traditional water quality parameters and a standardized fouling index: the MFI-UF (modified fouling index – ultrafiltration)

The specific objectives of this sub-project are:

- a. To characterize two natural water sources (Des Prairies River and Barron Lake) in terms of typical water quality parameters and fouling potential over an eight month period.
- b. To explore if there are any apparent relationships between the measured water quality parameters and the fouling potential of waters as measured by the MFI-UF.

3.2.2 Membrane pre-treatment evaluation

The main objective of this sub-project is to evaluate the fouling potential reduction achieved by automatic backwash filters as pretreatment for pressure driven membrane filtration. The fouling potential is quantified by means of a standardized fouling index (the MFI-UF).

The specific objectives of this sub-project are:

- a. Development of a bench scale filtration protocol that is representative of the particle removal by automatic backwash filters.
- b. Determination of the particle removal performance of fine screens and thread filters used in automatic backwash filters.
- c. Determination of the effect that pre-filtration has on the fouling potential of natural waters as measured by the MFI-UF.

3.3 Hypotheses

3.3.1 Seasonal water quality and fouling potential analysis

The working hypotheses are:

- a. The influence of water quality on MFI-UF is source-water dependent
- b. The MFI-UF is correlated to the presence of natural organic matter (as measure by the TOC, UV₂₅₄ nm and color).
- c. Turbidity is not well correlated to the MFI-UF

3.3.2 Membrane pre-treatment evaluation

The working hypotheses are:

- a. Pre-filtration will remove particles larger than the announced pre-filter porosity.
- b. The greater the particle removal, the greater the MFI-UF reduction will be.
- c. The formation of a cake layer on the surface of a filter will improve its particle removal performance while operating at a constant flowrate.

CHAPITRE 4 : PRELIMINARY RESULTS

The results presented in this chapter pertain to challenges met during the course of the laboratory work conducted that could be helpful or of interest to other researchers working with bench-scale evaluations of membrane fouling. The method used for calculating the MFI-UF is also discussed.

4.1 Membrane clean water permeability

As mentioned previously, the ultrafiltration membrane coupons that were used for the MFI-UF measurements were pre-conditioned prior to clean water permeability (CWP) determination in order to limit variability. Permeability is flux divided by operating pressure. Coupons were rejected if they had an initial permeability that varied by more than 20% from the manufacture quoted permeability, that is outside the range of $1.57 \times 10^{-7} - 2.33 \times 10^{-7} \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1} / \text{kPa}$ ($56 - 84 \text{ L m}^{-2} \text{ h}^{-1} / \text{bar}$) or if they failed to attain a stable normalized CWP (permeability deviation of less than 3%) during the last 1.5 hours of filtration. Total filtration run durations were between 3 to 4 hours.

Membrane details provided by the manufacturer are presented in table 4-1. The permeability measurements were done at 207 kPa (30 psi) and 20 °C while the conditions specified by the manufacturer were 207 kPa (30 psi) and 25 °C. The normalization of the flux measurements to 25 °C was done using equation 4-1:

Table 4-1: Membrane details

Material	MWCO (approximate)	Normalized water flux_a	Rejection of 20kDa PEG markers_a
Polyacrylonitrile	10 kDa	70 lmh/bar	95%

^a The normalized flux was measured with non-fouling RO permeate at 30 psi and 25 °C.

$$J_s = J_m \left(\frac{\Delta P_s}{\Delta P_m} \right) \times 1.024^{(T_s - T_m)} \quad (\text{Equation 4-1})$$

Where

J_s = flux corrected to standard pressure and temperature ($\text{L m}^{-2} \text{h}^{-1}$)

J_m = measured flux ($\text{L m}^{-2} \text{h}^{-1}$)

P_s = standard pressure (30 psi)

P_m = measured pressure (psi)

T_s = standard temperature (25 °C)

T_m = measured temperature (° C)

(Howe et Clark, 2002a)

Thirty-two membrane coupons were used for the seasonal water quality variation investigation and nine membrane coupons were used for the pre-treatment performance evaluations, all coupons were cut from the same 1 m^2 membrane flat sheet. The results presented here do not include the membrane coupons that were rejected due to non conformity to the criteria specified above.

The distribution of the permeability was found to be Gaussian with a Shapiro – Wilk W of 0.966 and p value of 0.253 (figure 4-1). The mean of the distribution was $1.74 \times 10^{-7} \text{ m}^3 \text{ m}^{-2} \text{h}^{-1} / \text{kPa}$ ($62.71 \text{ L m}^{-2} \text{h}^{-1} \text{ bar}^{-1}$) at 25 °C and the standard deviation was 1.55×10^{-8}

$\text{m}^3 \text{m}^{-2} \text{h}^{-1} / \text{kPa}$ ($5.59 \text{ L m}^{-2} \text{h}^{-1} \text{bar}^{-1}$). It should be noted that the mean permeability was 10% lower than manufacturers announced permeability.

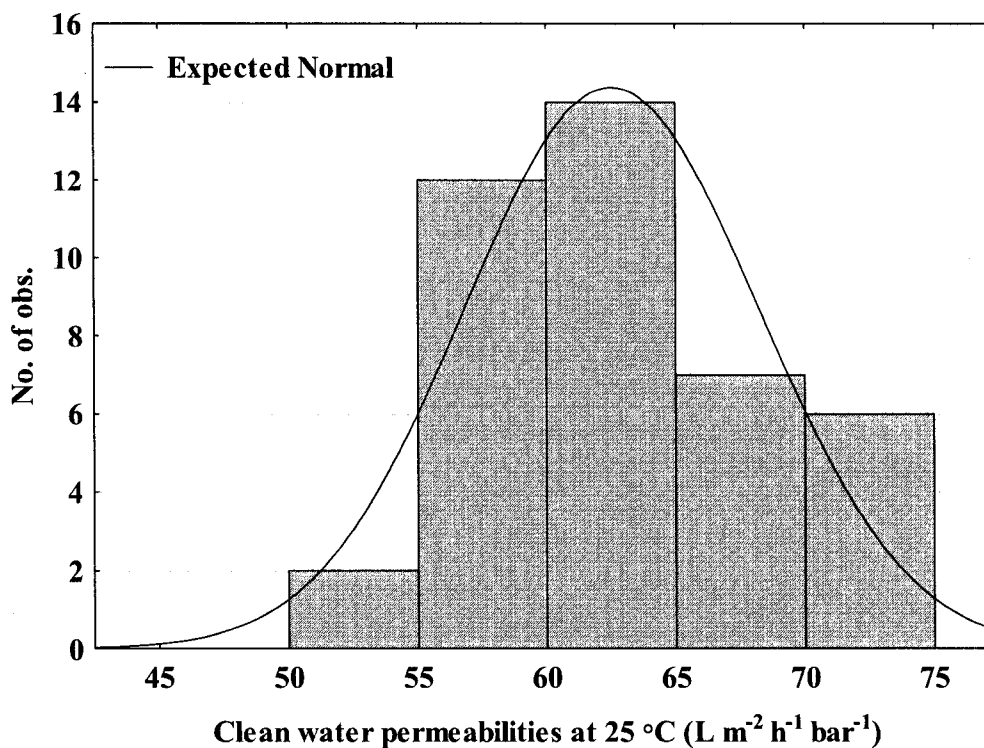


Figure 4-1: Clean water permeability distribution

4.2 Pre-filtration with laboratory filters: a cautionary tale

Prior to the decision to use actual pre-filter samples, the use of laboratory disc filters was considered to determine which particle size range played the most important role in fouling potential. Polypropylene 47 mm filters (Millipore Corporation) of varying

porosities (0.6 μm , 1.2 μm , 2.5 μm , 5 μm , 10 μm , 30 μm) were used to pre-filter raw water samples from the Des Prairies River. Upon filtration, it was noticed that the turbidities of the filtrates were unusual, with the 1.2 μm filtrate having a higher turbidity than the 30 μm filtrate. Filtrations were then repeated to verify that it was not human error that was the source of the discrepancies. Particle counts were carried out on these filtrate samples, results are presented in figure 4-2. It was thus found that the particle removal by the filters presented inconsistencies. The filters with the best overall particle removals were the 2.5 μm filter (1.64 logs) followed by the 5 μm (1.17 μm logs), the 30 μm (0.88 logs) and the 10 μm (0.81 logs) filters. Surprisingly, the filters that were the least successful at removing particles were the 0.6 μm (0.45 logs) and 1.2 μm (0.30 logs) filters.

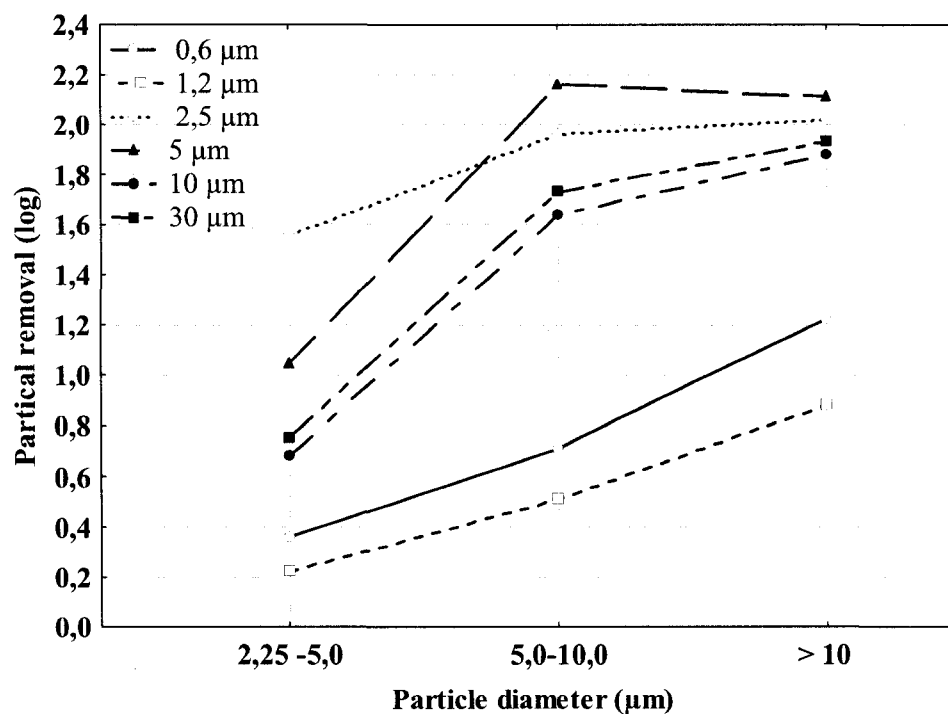


Figure 4-2: Particle removal by laboratory filters

Since great attention had been taken at the time of the filtrations to assure that there was no mix up of the filters, other possible sources of error were considered. Tests were performed to determine if there was a problem with the particle counter cell/tubing cleaning protocol and to verify if particles were released by the filters. Particle counts of ultrapure water run through the cell and tubing between sample filtrations (ultrapure) and of ultrapure water filtered through the 0.6 μm filter (ultrapure – 0.6 μm filter) were carried out, results are shown in figure 4-3. These counts are compared with raw water sample counts and with counts of raw water filtered through a 0.6 μm filter (Raw water – 0.6 μm filter).

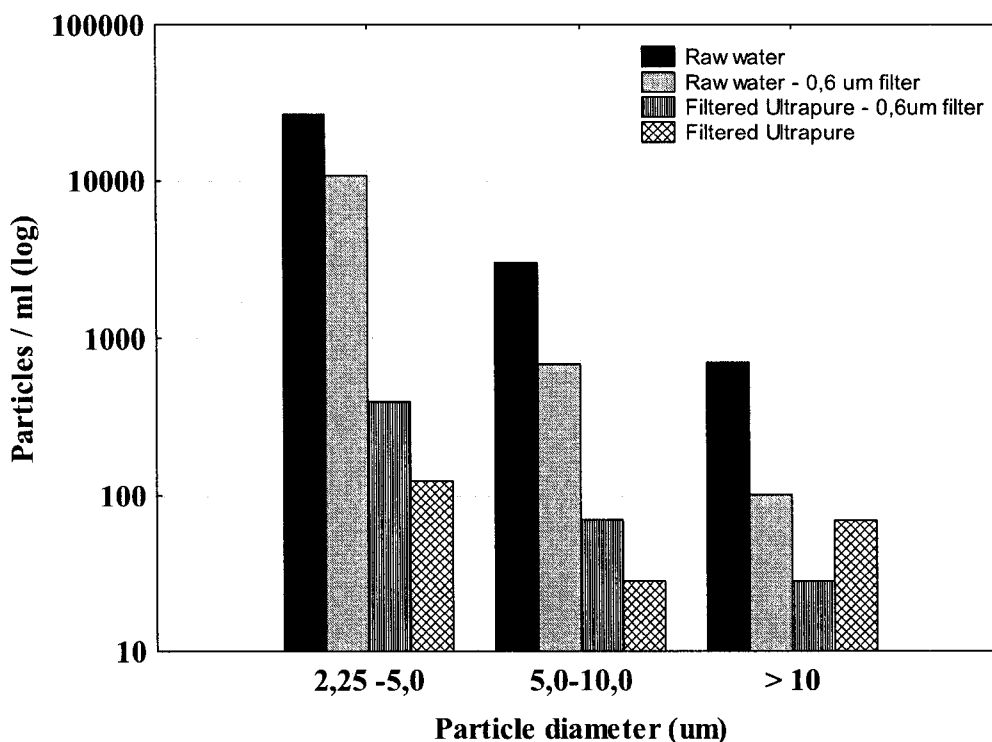


Figure 4-3: Particle counts for identification of error source

The total particle concentrations for the different samples are presented in table 4-2 below. The ultrapure water run through the system after the filtration of the raw water sample followed by the usual cleaning of the cell had a negligible particle concentration in comparison with the other concentrations in question. Therefore the problem did not appear to be improper cleaning. There were some particles released by the 0.6 μm filter, it was negligible compared to the particle concentrations found in the raw water sample and in the raw water sample filtered through 0.6 μm filter. Particle release by the filters did not explain the inconsistencies in particle removal.

Since all the possible sources of error that were considered as likely causes for the unexplained filter performances were tested and were not found to satisfactorily explain the observed inconsistencies, it was concluded that it was most likely a manufacturing defect that was responsible for the incoherent results.

Table 4-2: Total particle concentration for identification of error source

Sample	Raw water	0.6 μm filtrate (raw water)	0.6 μm filtrate (ultrapure water)	Ultrapure water
Total concentration (particles / ml)	30 299	15 636	494	221

4.3 MFI-UF calculation method

As described in the literature review, the MFI-UF is defined as the slope of the linear portion of the t/V vs V graph. In order to accurately measure the MFI-UF, it was first necessary to determine where the linear section of the graph began. This was done by means of the coefficient of variance, which is the standard deviation of a sample divided

by its average. The linear section began at the lowest cumulative filtration volume for which the instantaneous MFI-UF measurements up till the end of the experiment (7 hours) had a coefficient of variance that was 10% or lower. The samples MFI-UF was then measured as the slope of this section.

CHAPITRE 5 : BENCH-SCALE ASSESSMENT OF MEMBRANE PRE-TREATMENT AND SEASONAL FOULING POTENTIAL VARIATIONS

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5.1 Abstract

Fouling is widely recognized as an important challenge to the widespread use of membrane filtration technologies in water and wastewater treatment. Unfortunately fouling by natural waters is complex and its mechanisms are not presently well understood. Pre-treatment of feed water is part of a successful fouling control strategy. The aim of this study was to gain a better understanding of membrane fouling by characterising two natural waters seasonally using the modified fouling index - ultrafiltration (MFI-UF) and other various traditional water quality parameters (turbidity, NOM, UV₂₅₄, etc). In addition, fouling prevention by automatic backwash filtration was studied at a bench scale using samples of filter screens and thread filters. Apparent relationships between typical water parameters and fouling potential were explored through partial least square regression and pre-treatment performance was evaluated via particle counts and MFI-UF measurements. Results showed that it is not feasible to identify foulants using traditional water quality parameters as they lack the precision to specifically describe the actual foulants and that pre-treatment that removes large particles (> 2µm) only is ineffective at reducing fouling potential.

Keywords: membrane, fouling, pre-treatment, MFI-UF, particles

5.2 Introduction

Nanofiltration is a versatile solution for many of the water quality issues that drinking water producers are faced with today. NF membranes can be used for the removal of NOM (natural organic matter) [1-4], cyanotoxins [5-7], pathogenic organisms [8] and endocrine disrupting compounds (EDC) [9]. Membrane systems are particularly interesting for smaller systems due to their small foot-print, high level of automation and relative independence from chemical additives. However, one of the most critical challenges to a more widespread application of membrane technology remains membrane fouling [10].

Understanding membrane fouling is necessary for the advancement of membrane technologies and numerous studies have been conducted to gain a greater knowledge of the physical and chemical factors controlling fouling of NF membranes [11-13]. [13-16]. However, many of these studies have focused on observing individual fouling mechanisms either by using synthetic waters [15,16] or by fractionating natural waters into different components [12-14] before studying each component individually. This does not provide a comprehensive picture of fouling since there is often an overlap between the fouling types [17] and since synthetic waters do not necessarily behave the same way natural waters do [18].

Estimation of the fouling potential of feed water based on traditional water quality parameters has not been successful [19-21]. Fouling indexes have been developed to meet the need for a fast and accurate measurement of fouling. Of the many fouling indexes proposed in the literature [22-26] the MFI-UF (modified fouling index – ultrafiltration) [27] was found to be one of the most interesting options. This index was developed based on cake filtration, which is the fouling mechanism thought to dominate in high pressure membrane fouling [28].

Measuring fouling potential is essential to gauging the performance of physical and/or chemical pre-treatments which are integral parts of a complete fouling control strategy. While there is a wide variety of potential pre-treatment options, the schemes most often proposed in the literature to control fouling in high pressure membrane systems [29-31] are methods that require either continuous chemical addition, construction of important infrastructure or additional low pressure membrane systems upstream. These constraints are often prohibitive for smaller systems. A treatment technique which does not have these constraints is automatic backwash filtration. Successful operation of NF systems that implemented this pre-treatment technology has been reported [32,33] but results did not elucidate the actual improvements made by introducing the pre-treatments.

The present study aims at gaining a better understanding of fouling by natural surface waters and its prevention through physical pre-treatment in order to facilitate the use of spiral wound high pressure membrane filtration systems in small communities. MFI-UF measurements and various traditional water quality parameters were used to describe the fouling potential of raw natural waters over an eight month period. Meanwhile, laboratory filtrations using samples of filters screens and thread filters were used to simulate the performance of automatic backwash filter technology. Industrial filter samples were used rather than laboratory grade filter discs that are more commonly used in bench scale experiments. This was done to gain a better idea of the performance of full scale filters. Pre-treatment performance was evaluated via particle counts and MFI-UF measurements.

5.3 Materials and methods

5.3.1 Feedwaters

Two natural surface waters were used in this study. Analyses of water quality variations were carried out on samples taken from the raw water intakes of the drinking water treatment plants of the city of Laval (source water Des Prairies River) and the city of Lachute (source water Barron Lake) from February to October 2007. Both locations are situated in the Province of Québec, Canada. Only the more heavily particle charged Des Prairies River feed water was used in the pre-treatment analysis section as it represented the greater challenge in terms of particle loading. The samples were stored in a dark cold room (4°C) on arrival and were kept there until the experiments were performed; samples were conserved for a maximum of 24 days.

5.3.2 MFI-UF measurements

MFI-UF theory

As mentioned previously, the MFI-UF is a fouling index which is used to measure fouling potential based on the cake filtration theory. This theory states that the increase in hydraulic resistance is caused by the formation of a cake layer formed by particles retained on the external membrane surface. This increase in resistance is proportional to the cumulated volume of filtered water, V [34]. Thus the MFI-UF is a function of the concentration and nature of the particles in the feed water. The MFI-UF is measured at a constant pressure gradient through an UF membrane and it corresponds to the slope of the linear portion of the t/V vs V curve (see equation 5-1) where t is the filtration time.

Measurements were taken at standard conditions; 20° C and 207 kPa [27]. If the actual conditions departed slightly from these reference conditions, the MFI-UF values were standardized using equation 5-2.

$$\frac{t}{V} = \frac{\eta R_m}{\Delta P A_m} + \left(\frac{\eta C_b \alpha}{2 \Delta P A_m^2} \right) V = \frac{\eta R_m}{\Delta P A_m} + (MFI - UF) V \quad (\text{Equation 5-1})$$

$$MFI - UF_{std} = \frac{\eta_{20}}{\eta_T} \left(\frac{\Delta P}{207 \times 10^3 \text{ Nm}^{-2}} \right) MFI - UF_{measured} \quad (\text{Equation 5-2})$$

Where:

t is filtration time (s)

V is the permeate produced (m³)

ΔP is the transmembrane pressure (N m⁻²)

η is the dynamic viscosity of water (N s m⁻²)

A_m is the membrane area (m²)

R_m is the membrane resistance (m⁻¹)

α is the specific cake resistance (m kg⁻¹)

C_b is the particle concentration (kg m⁻³)

Measurement set-up

The MFI-UF was measured using the bench scale set-up presented in figure 5-1. The feed water samples were placed in a stainless steel reservoir and maintained under constant pressure by means of a nitrogen gas tank whose pressure was controlled by a pressure regulator (Tescom) with an accuracy of ± 3.5 kPa (0.5 psi). The pressure was monitored with a digital pressure gauge (Ashcroft) with a full scale of 414 kPa (60 psi), a

resolution of ± 0.07 kPa (0.01 psi) and an accuracy of ± 2.1 kPa (0.3 psi). The membrane filtrations were conducted in unstirred dead-end filtration cells with an internal volume of 50 ml (Amicon-Millipore Corporation). Permeate flux was calculated from cumulative gravimetric mass measurements which were continuously recorded using an electronic balance (Sciencetech) with a full scale of 3000 g, a readability of 0.01g and a repeatability of 0.01 g. Data was collected automatically every 5 minutes. Experiments were carried out at room temperature ($20 \pm 3^\circ$ C) and at 207 kPa (30 ± 1 psi), temperature and pressure were noted at the beginning and the end of every experiment. Pressure was adjusted at the beginning of the experiment and was not significantly altered during the rest of the experiment.

Membrane preparation

The membrane used for all MFI-UF measurements was a flatsheet polyacrylonitrile (PAN) membrane with a MWCO of approximately 10 KDa (Sepro Membranes, USA). This membrane was chosen as it was the membrane found which was most similar to the reference membrane recommended for MFI-UF measurement by [28]. A new membrane coupon was used for every experiment in order to avoid the effects of irreversible fouling. Conditioning has been shown to produce more homogenous fluxes in membranes with values closer to those quoted by manufacturers [35]. Therefore membrane coupons were soaked in laboratory grade water (Milli-Q) for a minimum of 12 hours previous to the experiments to remove any preservation agents. Membranes were then stabilised by pressurized filtration first at 310 kPa (45 psi) for 15 minutes and then at 207 kPa (30 psi) until stable clean water flux was attained again using laboratory grade water (between 3 and 4 hours). In order to insure a certain homogeneity, coupons were rejected if they had an initial flux which varied by more than 20% from the pure water permeability quoted by the manufacturer $1.94 \times 10^{-7} \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1} / \text{kPa}$ ($70 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) or if they failed to attain a stable clean water flux (flux deviation of less than 3%) during the last 1.5 hours of filtration. Typically this second criteria was verified after 3 hours and, if

had not been attained, the filtration was allowed to continue up to 4 hours after which, if the criteria had still not been attained, the coupon was rejected.

5.3.3 Benchscale pre-treatment

In order to develop an adequate filtration protocol which was representative of particle removal by full scale automatic backwash filters at a laboratory scale, samples of filters screens and textile filters employed by Amiad Filtration Systems (www.amiadusa.com) in full scale installations were used. The automatic backwash filter options proposed by this company were interesting since they included a thread filter technology with filtration degrees that went down to 2 μm , which was by far the finest particle removal available found for self-backwashing filters. Moreover an article published by [32] described the successful use of this technology upstream of NF membranes. Even though laboratory filters have been previously used in bench-scale studies to model the behaviour of different treatment technologies [12,36], it is highly unlikely that these filters will perform in the same manner as filters produced for large scale applications.

Pre-filtration – Role of filter porosity

Four pre-filters of different porosities and different technologies were used in the study, 25 and 10 μm stainless steel screen filters and 3 and 2 μm thread filter cassettes. The 10 and 25 μm stainless steel screens were cut into 40 mm diameter coupons with an active filtration area of 11.4 cm^2 and placed in the pre-filter housing of a stainless steel filter holder (Millipore Corporation) in order to filter the sample at 151.7 kPa (22 psi) which was the minimum operating pressure for the full scale filter (figure 5-2). Due to the high flux through the screens and the small volume of filtrate necessary ($\sim 2\text{L}$), the duration of the filtration run was very short (a matter of seconds) and it was not practical to record the variation of flux throughout the run. The first liter that was filtered through the screen was discarded and the second liter was collected for MFI-UF measurements and

particle counting. The 2 and 3 μm cassette filters, which are made of high strength polyester thread wound around a plastic cassette with an active filtration area of 102 cm^2 , were first soaked for 10 minutes in the water sample that was to be filtered and filtrations were then performed at a constant flowrate of 500 mL/min (flux of $2.95\text{ m}^3\text{ m}^{-2}\text{ hr}^{-1}$) as suggested by the manufacturer. The initial and final differential pressures were 9.7 kPa (1.4 psi) and 16.5 kPa (2.4 psi) respectively for the $2\mu\text{m}$ cassette and 3.5 kPa (0.5 psi) and 9.0 kPa (1.3 psi) for the $3\mu\text{m}$ cassette. The pressure of the influent was measured and the outflow was at atmospheric pressure (figure 5-3). As done previously, the first liter filtered was discarded and the second liter was collected for MFI-UF measurements and particle counting. All experiments were conducted at room temperature.

Pre-filtration – role of cake formation

An experiment with a $2\text{ }\mu\text{m}$ cassette filter was done to investigate the role that cake formation plays on pre-filter performance in terms of particle removal and MFI-UF reduction. The filter cassette was soaked for 10 minutes in the raw sample and the filtration was done at a constant flowrate of 500 mL/min (flux of $2.95\text{ m}^3\text{ m}^{-2}\text{ hr}^{-1}$). The pressure differential across the cassette was initially 4.1 kPa (0.6 psi) and by the time a cumulate volume of 12 L had been filtered, it had surpassed 103 kPa (15 psi) which was the full scale on the pressure gage on the set-up. Samples of filtrate were collected at different specific volumes (volume filtered / cassette filter area) during the filtration run; they were collected at $0.2\text{ m}^3/\text{m}^2$, $0.7\text{ m}^3/\text{m}^2$, and $1.2\text{ m}^3/\text{m}^2$ for particle counts and MFI-UF measurements.

5.3.4 Additional water quality analysis

Several analytical methods were used to measure traditional water quality parameters to complement the information on fouling potential. Temperature and pH (AB15 Accumet basic, Fisher Scientific) were measured but were not included in the data analysis as they

did not vary significantly for either source, pH values varied from 6.9 to 7.6 for Barron Lake and from 7.2 to 7.9 for the Des Prairies River, temperature for both sources were between 19 and 22 ° C. Ultraviolet absorbance at 254 nm (UVA), color (Cary 100, Varian) and total organic carbon (TOC) (5310C Laboratory TOC analyser, Sievers) were measured to characterize the amount and nature of the organic matter in the samples. The ionic content of samples was characterised through conductivity (Conductivity Testr, Oakton instruments) and total, Fe, Mg and Ca concentrations (AAnalyser200, PerkinElmer). Particle counts (DPA 4100, Brightwell technologies) and turbidity (2100AN, HACH) were measured to characterize the particulate and particulate / colloidal content respectively. It should be noted that the DPA 4100 can only measure particles in the 2.25 – 300 µm range. The specific ultraviolet absorbance (SUVA) was calculated based on the TOC concentrations and UV_{254} absorbance.

5.3.5 Data analysis – Seasonal water quality variations

Principal component analysis (PCA) and partial least squares (PLS) regression were done on the data from both sampling sites, first in conjunction and then separately. PCA is a multivariate exploratory technique that is useful for the classification of variables and observations and thus for data interpretation [37]. PLS regression is a linear regression method that can be used in situations where traditional multivariate methods cannot be applied such as when there are few observations and many collinear variables [37,38]. This was the case of the data in this study with 10 predictor variables and 16 observations per water source. Statistical analyses were performed with Statistica 7.1 [37].

5.4 Results

5.4.1 Seasonal water quality variations

As mentioned previously water quality parameters were measured from February to October 2007 at two different sampling locations throughout different seasonal conditions (precipitation, snowmelt, temperature variations, etc.); one source was a lake the other a river. The ranges of the values found for the quality parameter measured at Barron Lake and the Des Prairies River are presented in figures 5-4 to 5-6. As expected the lake water had a different quality profile than the river water. These differences will be discussed in greater details further on in this section.

It should be noted that particles with diameters between $2.5 - 3 \mu\text{m}$ represented the most important size fraction of the total counts for both for Barron Lake and the Des Prairies River. The average percentage of particles that fell within this size range were $63\% \pm 6\%$ and $68\% \pm 4\%$ of total counts for the two sources respectively. Particles larger than $10 \mu\text{m}$ were consistently the least important fraction with averages of $5\% \pm 4\%$ and $3\% \pm 1$ for Barron Lake and the Des Prairies River. Particles larger than $30 \mu\text{m}$ represented on average less than 0.5 % of the total particle concentration for both sources.

A PCA was carried out using the observations of traditional water quality parameters collected at both sites (with the exception of the MFI-UF) to better gauge if it was appropriate to analyse the results from both sampling sites together. The PCA results showed that the samples collected from the different sources varied predominantly along two different factors which are graphically represented by the vertical and horizontal axes (figure 5-7). A PLS regression was done to further investigate the role water source plays in result interpretation. It was found that sampling location was the variable with the

greatest influence on the model produced by this method. Therefore the statistical analyses that followed were done separately for each of the sampling locations.

PCAs for each of the sources elucidated the multicollinearity between several of the variables measured. TOC, UV_{254} , color and turbidity all varied in the same direction along the same factor for the samples collected at Barron Lake (figure 5-8), all parameters had peak values during the beginning of June and end of October, (see figure 5-9). Turbidity, conductivity, Ca, Mg and Fe concentrations as well as particle counts all varied in a similar manner for the samples collected at the Des Prairies River (figure 5-10), these variables had peak values mid April, during the period of snowmelt (see figure 5-11). Color, UVA and SUVA also varied similarly at the Des Prairies River (no marked peaks during the sampling period).

In general the parameters associated with particulate and colloidal matter (turbidity and particle counts) were significantly higher for the river samples than for the lake samples. The parameters associated with ionic content (Fe, Mg, Ca) had slightly higher values for the river water samples and the organic matter parameters had comparable values for both sampling locations, but lake samples tended to have slightly more marked minimum and maximum values. Surprisingly, the ranges of the MFI-UF values were nearly the same for both water sources despite the significant differences in the signatures of the two waters (figures 5-4 to 5-6).

A PLS regression was done for each of the source waters in order to investigate any apparent relationships between the MFI-UF and the traditional water quality parameters that were measured. The models produced were able to predict MFI-UF values to coefficients of determination (R^2) close to 0.90 for both sampling sites. For the model built from measurements at Barron Lake the variables with the greatest influence on the variability of the MFI-UF were respectively turbidity, TOC and Fe concentration. Color and UV_{254} absorbance also played a role but to a somewhat lesser extent. For the model built from the Des Prairies River monitoring, the variables that were the most influential

on the prediction of the MFI-UF were respectively turbidity particle counts and Fe concentrations and to a slightly lesser degree conductivity and Mg concentrations. Figures 5-9 and 5-11 present this information graphically.

5.4.2 Pre-treatment

Both the effects of pore size and cake formation on pre-filter particle removal and fouling potential reduction were investigated. Traditional water quality parameters other than particle counts and turbidity are not reported since the filters used were expected to affect parameters measuring particle content only (the minimum porosity studied was 2 μm). The UV_{254} , color, and conductivity of filtrate samples from the 2, 3 10 and 25 μm filters were measured as indicators of changes in NOM and ionic content. No appreciable changes were noted (data not shown).

Effects of pre-filter porosity on MFI-UF and particle counts

As mentioned previously four pre-filters (rated 2 μm , 3 μm , 10 μm and 25 μm) were used to determine the role pre-filter porosity plays on MFI-UF reduction and particle removal. The pre-filters ability to remove particles of different diameters was evaluated (figure 5-12). The 2, 3 and 10 μm filters were found to be capable of removing 74%, 82% and 66% of particles with diameters greater than the announced pore size respectively. The results for the 25 μm filter are not presented as the concentration of particles of diameters greater than 25 μm in the raw water was negligible (< 50 particles /ml). It is also important to mention that the concentration of particles greater than 10 μm in diameter in the filtrate were also extremely low (150 particles /ml).

Pre-filters reduced turbidity marginally, even for at lowest porosity (2 μm). The raw water sample used for this series of experiments had a turbidity of 3.12 NTU while the 2

μm , 3 μm , 10 μm and 25 μm filtrates had turbidities of 2.36 NTU, 2.55 NTU, 2.66 NTU and 2.67 NTU respectively. The filtrations were found to have little to no effect on the MFI-UF values (figure 5-12); variations that are less than 10% are considered as marginal [31], and all measurements were found to be within $\pm 10\%$ of MFI-UF of the raw water sample (30 000 s/L^2).

Effects of cake layer formation on pre-filter on MFI-UF and particle counts

The effects that the formation of a cake on a 2 μm cassette filter has on MFI-UF and particle counts are presented in figure 5-13. The removal of particles with diameters greater than 2.25 μm decreased slightly from 89% to 87% to 83% as the filtration run progressed. The raw water sample used for this series of experiments had a turbidity of 9.6 NTU while samples collected at the specific volumes of 0.2 m^3/m^2 , 0.7 m^3/m^2 and 1.2 m^3/m^2 had turbidities of 4.70 NTU, 3.83 NTU and 3.34 NTU respectively. Although the removal of larger particles ($> 2.25 \mu\text{m}$) was declining, the improved reduction in turbidity probably reflects the higher removal of smaller particles due to the cake layer. Nevertheless, even though a cake layer formed visibly on the cassette during the filtration (figure 5-14), the MFI-UF of the samples taken at specific volumes of 0.2, 0.7 and 1.2 m^3/m^2 did not vary significantly from that of the raw water sample (33 000 $\text{s/L}^2 \pm 12\%$). Even though the variations are above the 10% threshold value, they are not significantly higher and cannot be classified as major variations.

5.5 Discussion

5.5.1 Water quality variation data

Observations from the two sampling locations varied distinctly along different factors in the PCA. This fact suggests that it is not possible to analyze data from different source

types together. This conclusion was also supported by the model produced by PLS regression using all observations from Barron Lake and the Des Prairies River in conjunction. The model featured sampling location as the variable that contributed the most heavily to explaining the variability of the fouling potential reduction.

Despite the different signatures that these waters had, the range of the MFI-UF values found were highly comparable with respective averages and standard deviations of 40 000 s/L² and 14 000 s/L² for Barron Lank and 41 000 s/L² and 14 000 s/L² for the Des Prairies River. This lead us to believe that there was an unmeasured parameter or parameters that were responsible for flux decline. While turbidity was found to be strongly correlated to the MFI-UF both for Barron Lake and Des Prairies River samples, the vastly different turbidity values associated with similar MFI-UF values suggest that it is most likely a specific fraction of the particulate/colloidal matter that is responsible for the fouling observed. The TOC concentration of the Barron Lake samples was strongly correlated to the MFI-UF while TOC concentration for the Des Prairies River samples was not found to be particularly relevant to the relationship developed to model fouling for that water source. It is possible again that a specific fraction of TOC which is responsible for fouling, and this fraction varied proportionately with total organic carbon concentrations for Barron Lake and did not for the Des Prairies River. This implies that, when speaking about fouling, traditional water quality parameters lack the precision to describe and quantify the foulants which are causing the flux reduction amongst different source waters. However, the high coefficients of determination ($R^2 > 0.90$) obtained using PLS regression for each source waters indicate that traditional water quality indicators could be used to easily follow seasonal variations in fouling potential once the signature of a specific source water has been adequately described.

As mentioned previously the focus of many of the studies presented in the literature has been to observe and dissect a single fouling phenomenon, often this by means of synthetic or fractioned waters which simplify the control of certain parameters. This kind

of approach has led to the identification of possible foulants such as hydrophilic, high molecular weight/colloidal NOM [13,14,39,40], calcium – NOM complexes [11,41], iron oxides [19] and inorganic colloid – NOM complexes [16]. However, this kind of approach does not typically consider the effects of the seasonal variations that can occur with a raw natural water source. Even in the case where these variations have been considered the number of samples used remained small [42].

The work presented here attempted to create a context within which fouling potential can be interpreted by considering two different raw water sources over a prolonged period of time. Also, the difficulties associated with the analysis of many correlated variables were addressed by using a linear regression method that circumvents many of the limitations of more traditional linear regression methods.

5.5.2 Pre-treatment

The samples of filtration screens / thread filters tested did not completely remove all particles larger than the porosities announced by the manufacturer, this incomplete particle removal by commercial cartridge filters has also been noted by other researchers [12]. However, the thread filters consistently removed more than 70% (≈ 0.5 logs) of particles of greater size than the announced porosities. The removal of particles with diameters greater than 2 microns did not appear to significantly affect fouling potential of the natural waters. This was illustrated by the little to no change in MFI-UF values of raw water samples and pre-filtered samples. This result is in concurrence with findings published by [43] who also found that larger particles played a negligible role in fouling.

MFI-UF changes due to pre-treatment reported in the literature are in the range of 1 log reduction [44,45] while the changes that were observed here were very close to 10%, a value which changes can be considered marginal considering that MFI-UF can vary on a log scale. It seems likely then that the culprits behind fouling are colloids which are in

the sub-micron range. Organic colloids in particular have been identified by several researchers as being strong contributors to the fouling of MF / UF membranes [46,47] as well as NF membranes [13,14,48,49]. Evidence suggesting certain inorganic colloids such as iron oxides have a linear correlation to fouling has also been presented [19].

It should be noted that the raw water samples that were used to evaluate the efficiency of the pre-filters had very few particles with diameters greater than 10 μm . While particles larger than 10 μm were consistently the least important size fraction for all samples collected from both the Des Prairies River and Barron Lake, they were still sometimes present in non-negligible numbers. The average concentration of particles in this size range for Barron Lake was 500 particles /ml while for the Des Prairies River it was 2 000 particles /ml. Thus 10 μm pre-filtrations would provide some protection against plugging of spiral wound modules with source water like the Barron Lake and the Des Prairies River. Filtration coarser than 30 μm however would most likely be ineffective since particles larger than this porosity were typically found in negligible concentrations.

The 5 μm particle removal that is a standard specification requested by membrane manufacturers protects the feed spacer in spiral wound modules from obstruction by larger particles during events of high particle loading, but its effects on the protection of the membrane itself from fouling are negligible for the two sources studied here as illustrated by the results presented.

The formation of cake layer at a constant flowrate and increasing differential pressure did not significantly improve the performance of the filter in terms of MFI-UF reduction or particle removal. The pre-filter effluent collected at different specific volumes did not have MFI-UF values that varied by important amounts ($\pm 13\%$) from the raw water sample. Indeed, the removal of large particles decreased as the filtration run progressed. This is possibly the result of the increasing shear forces that developed in the filter due to the reduction of the open pore area, an effect reflected by the increase in differential

pressure. Thus, the cake layer formed under constant pressure did not succeed at progressively reducing fouling potential or the number of larger particles by virtue of lower cake layer porosity.

As mentioned previously the majority of the studies presented in the literature have focused on the low pressure membranes, conventional treatment [29,50,51], biological filtration [52], ozonation [30] and a combination of the above [31] There is therefore no wealth of information regarding simpler pre-treatments schemes to compare the present results to.

Since colloids appear to play a key role in fouling potential, future works should focus on developing better methods for colloid quantification (possibly colloidal counts) and applying these methods to natural waters to better understand the role colloids play in fouling in conjunction with larger particles, specific NOM fractions and ionic parameters.

5.6 Conclusion

In conclusion, it is not possible to analyse data from different source types together since traditional water quality parameters lack the precision to accurately describe the actual foulants present in natural water samples. However if a single water is studied, and comparisons between sources are not made, these parameters can be used to model fouling events and easily follow seasonal variations in fouling potential associated with climate events (spring turnover, heavy precipitation, algal blooms, etc) which could be useful in planning membrane piloting schedules to agree with the time when a pilot would face the greatest challenge in terms of fouling potential.

The automatic backwash filter samples tested did not completely remove particles from the feed, but there was a minimum reduction of 0.5 logs of particles larger than the announced thread filter porosity. The formation of the cake layer on the pre-filter

improved neither the removal of particles nor the fouling potential reduction. The removal of large particles only ($> 2 \mu\text{m}$) has a marginal effect on fouling potential reduction, and it is suspected that colloids in the submicron range are responsible for the observed fouling.

Future works should focus on the quantification and qualification of colloids in natural raw waters, and the comparison of this colloidal signature to the fouling potential over time.

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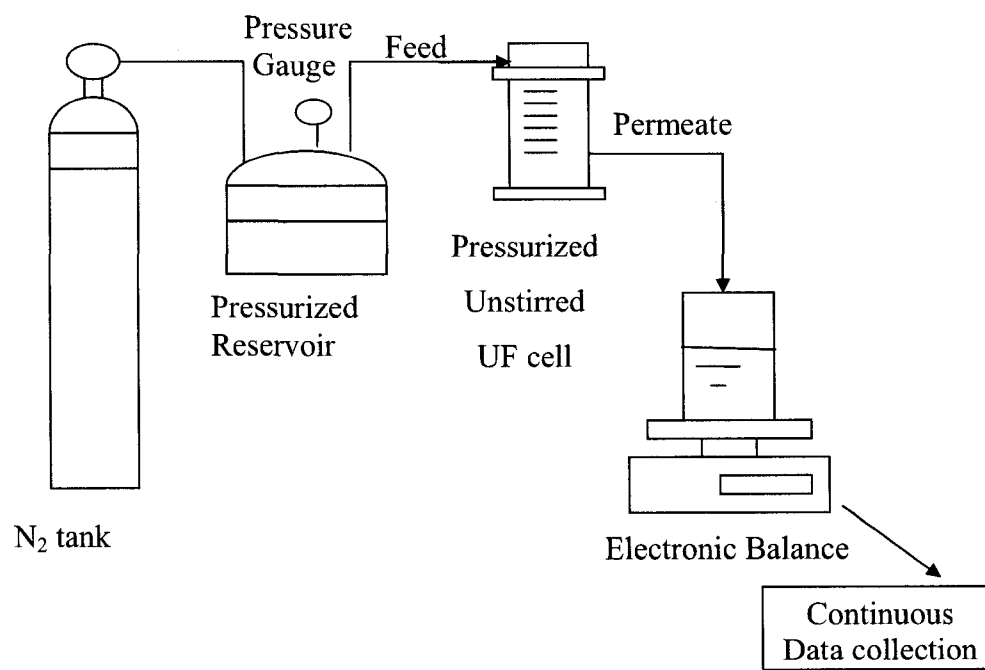


Figure 5-1: MFI-UF measurement set-up

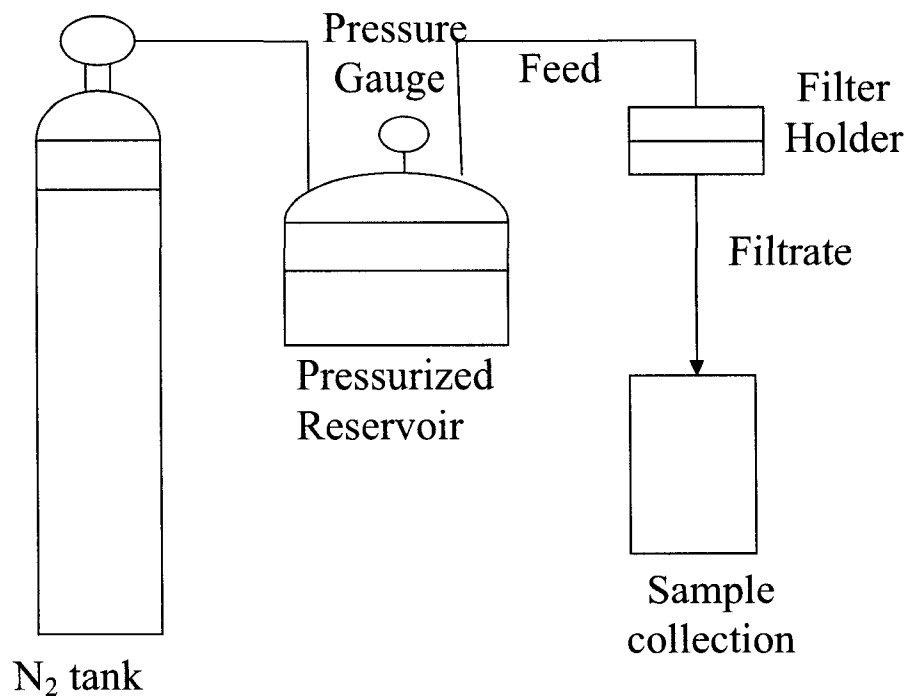


Figure 5-2: Screen filtration set-up

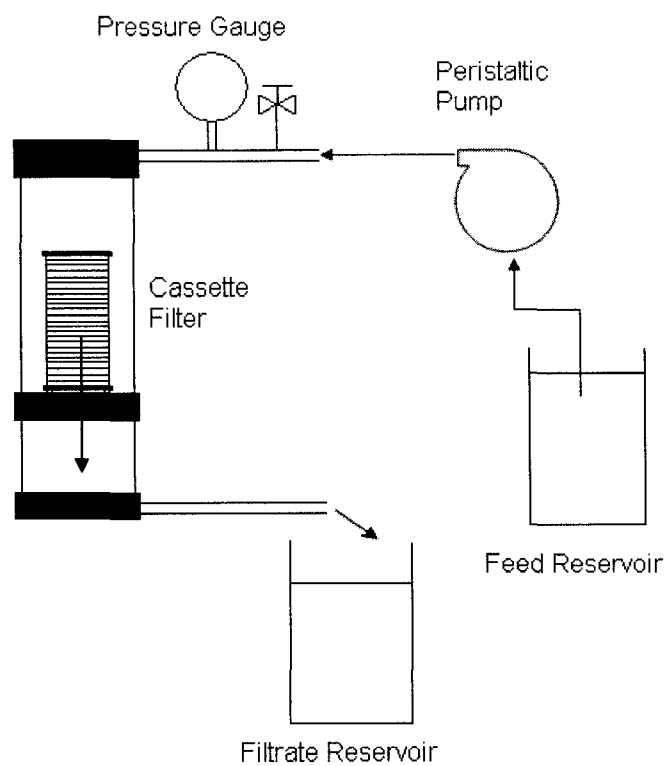


Figure 5-3: Thread filter set-up

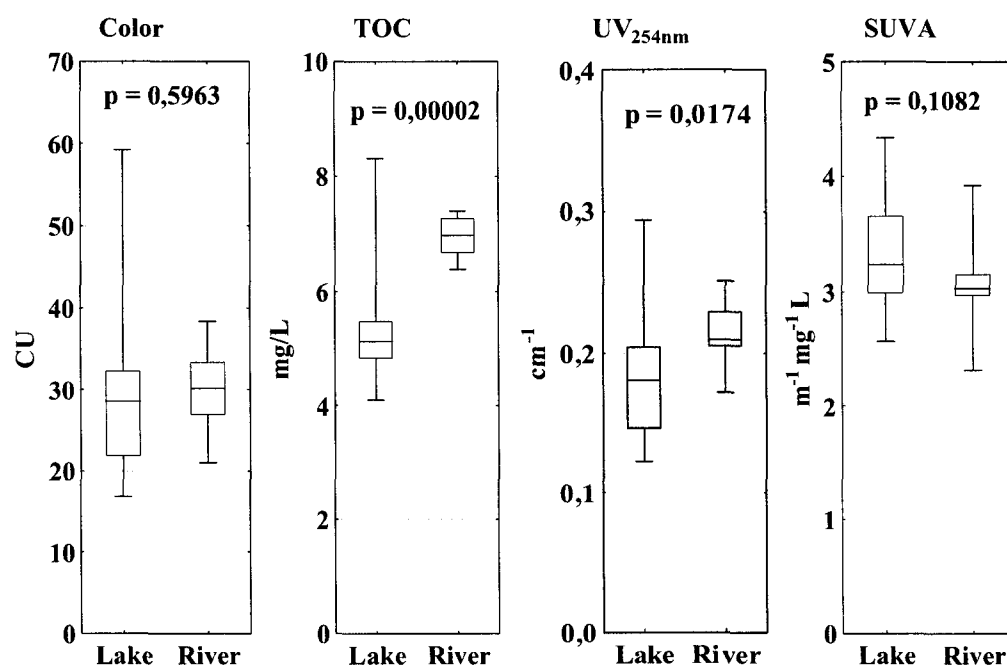


Figure 5-4: Range of NOM parameters measured during seasonal monitoring

Median 25%-75% Min-Max

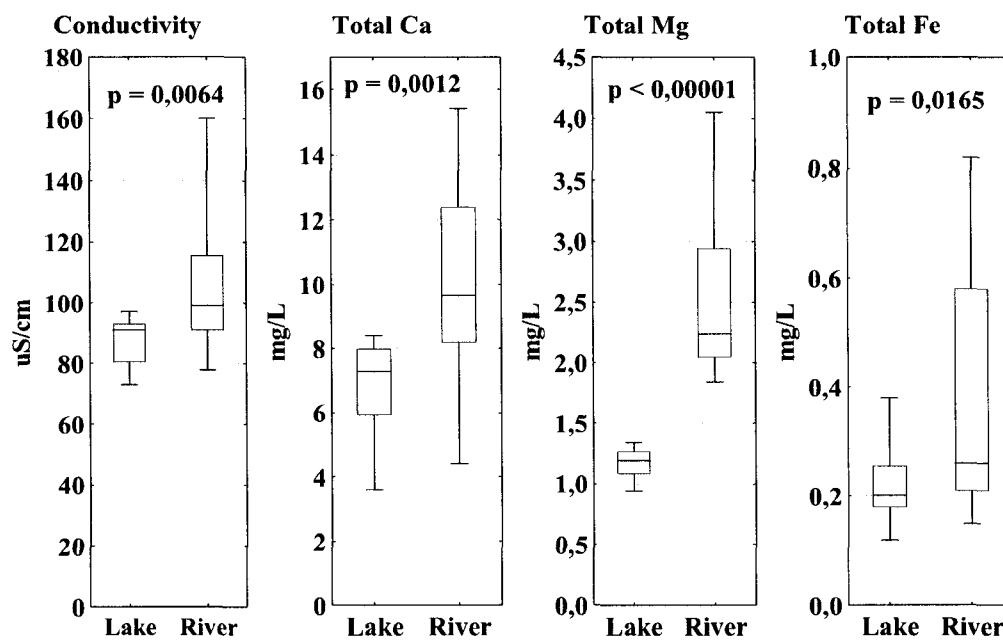


Figure 5-5: Range of ionic species measured during seasonal monitoring

Median 25%-75% Min-Max

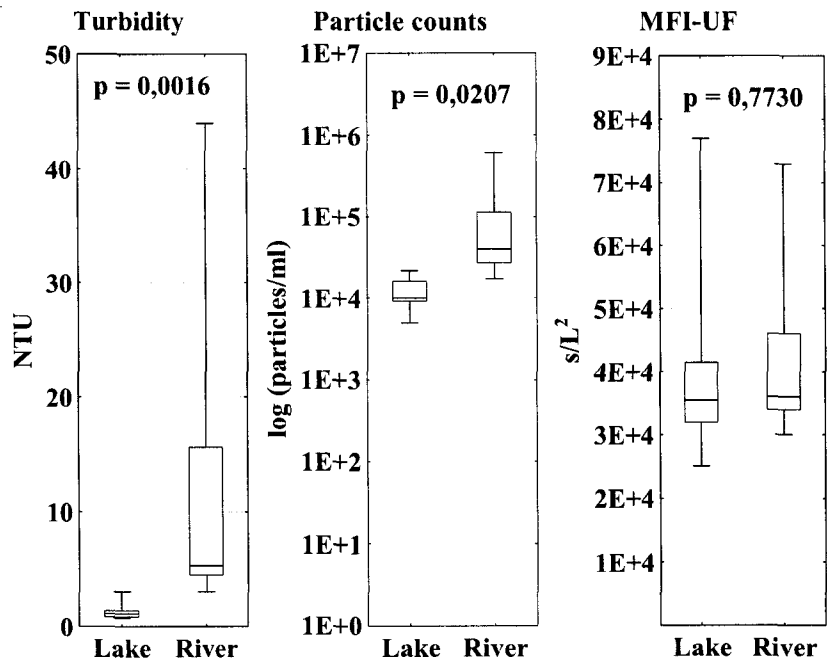
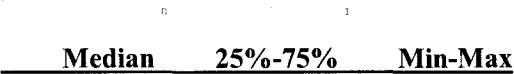


Figure 5-6: Range of solids parameters measured during the seasonal monitoring



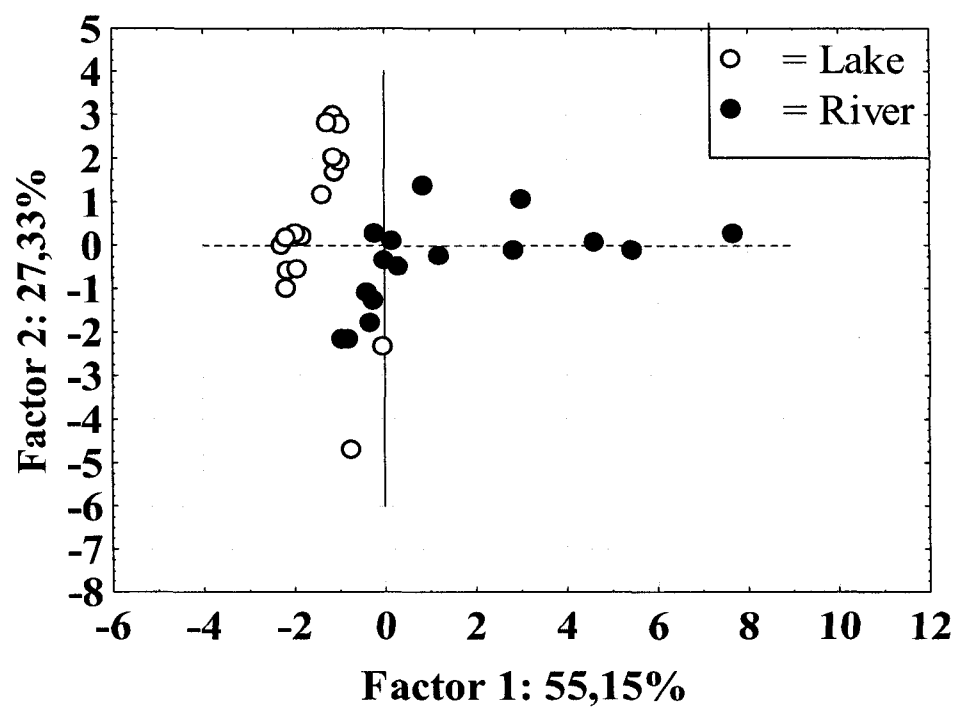


Figure 5-7: PCA case projection - all observations

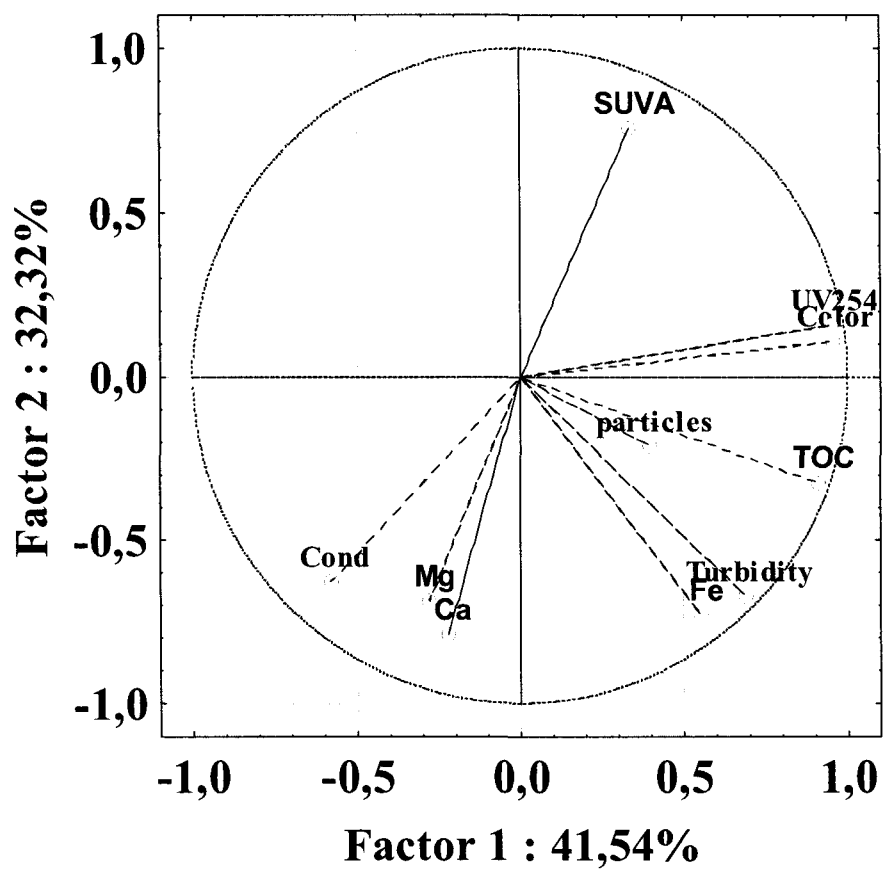


Figure 5-8: PCA variable projections – Barron Lake

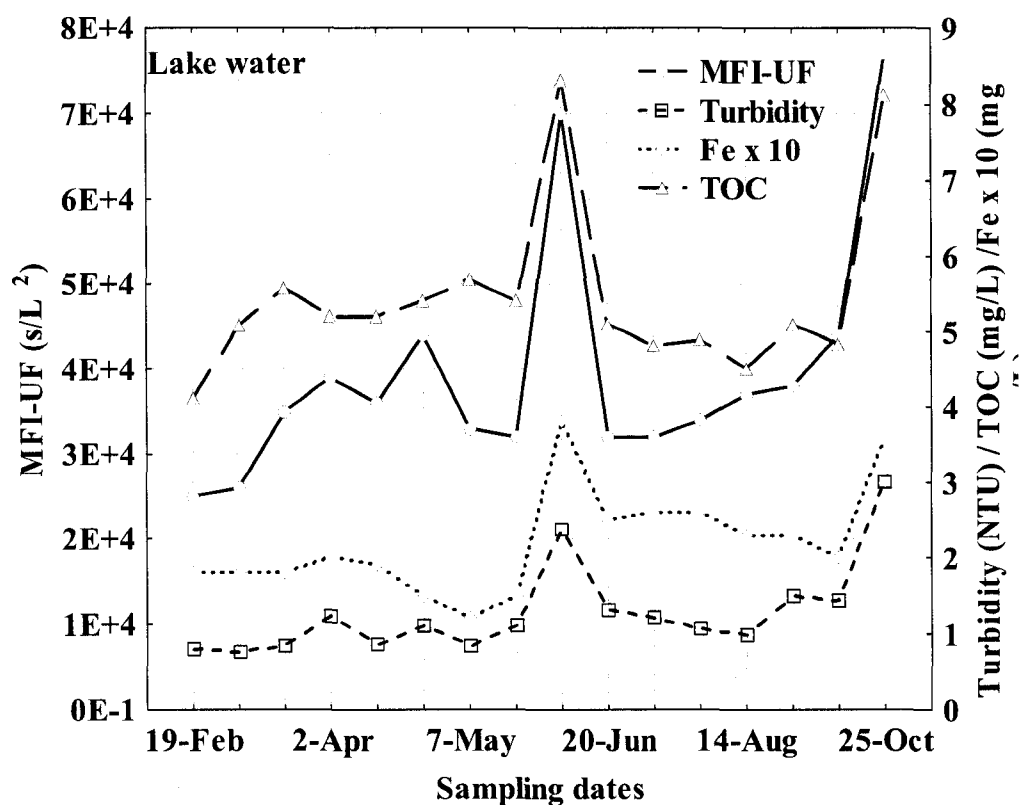


Figure 5-9: Barron Lake - fouling potential and related variables

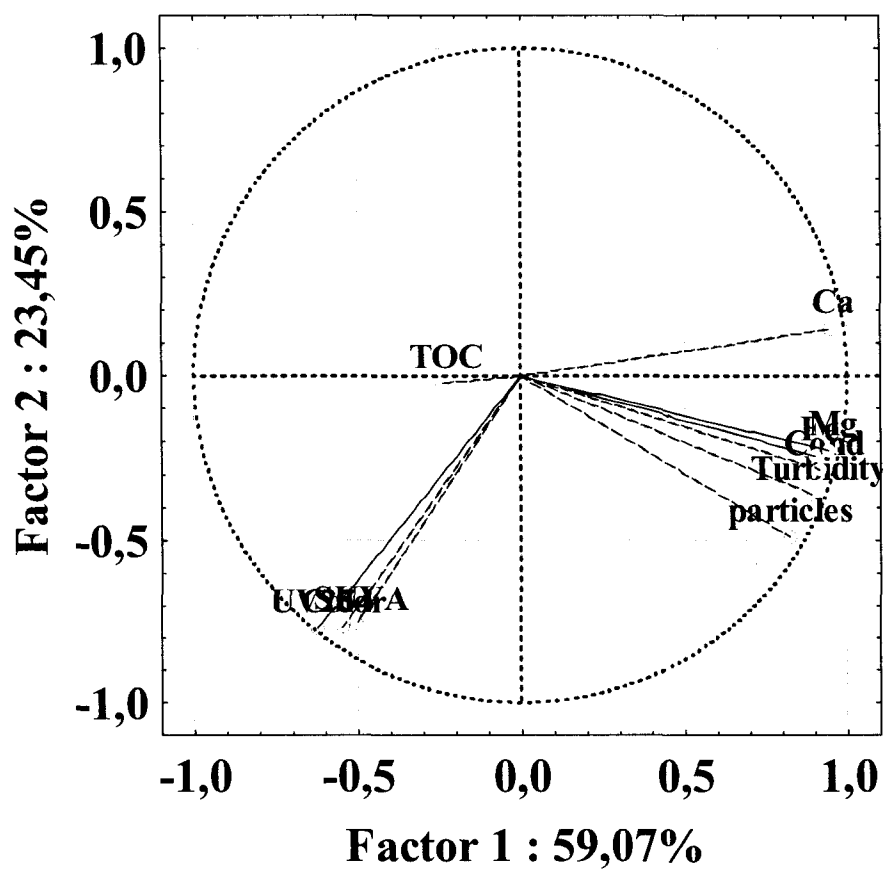


Figure 5-10: PCA variable projections - Des Prairies River

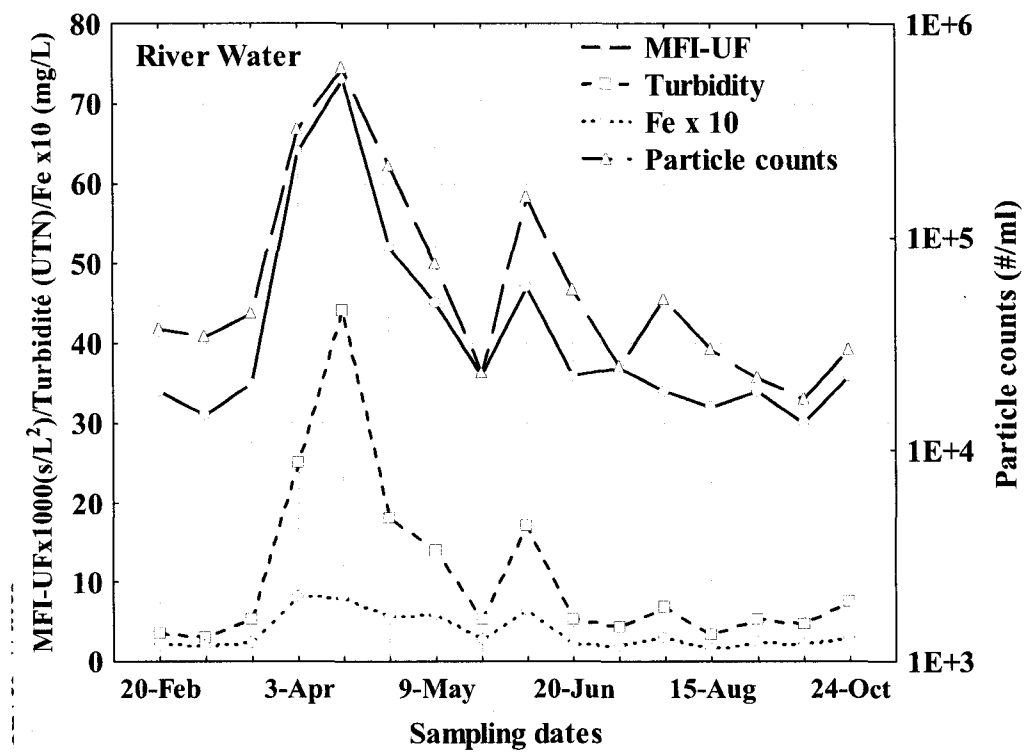


Figure 5-11: Des Prairies River - Fouling potential and related variables

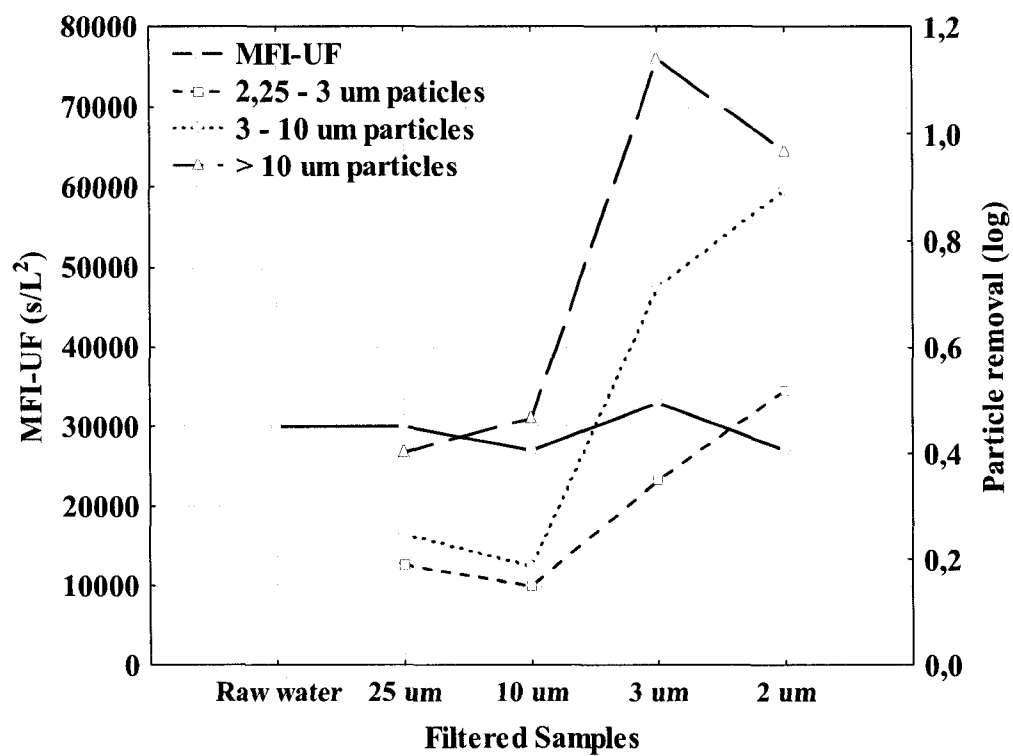


Figure 5-12: Pre-filter performance pore - size effects

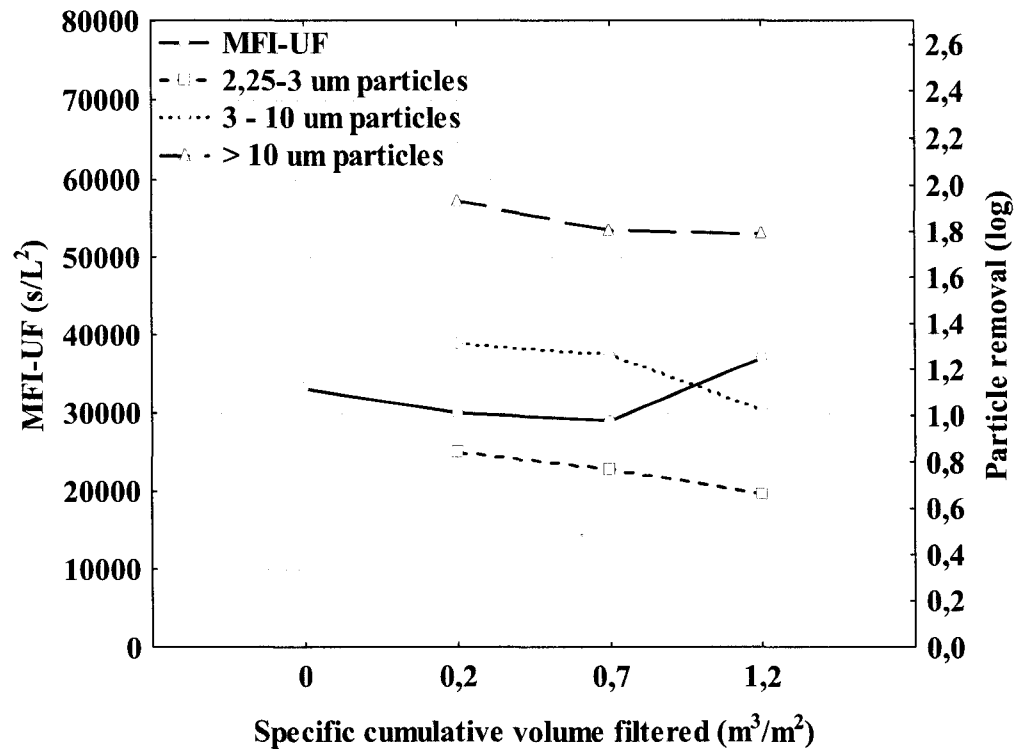


Figure 5-13:Pre-filter performance - cake layer effects

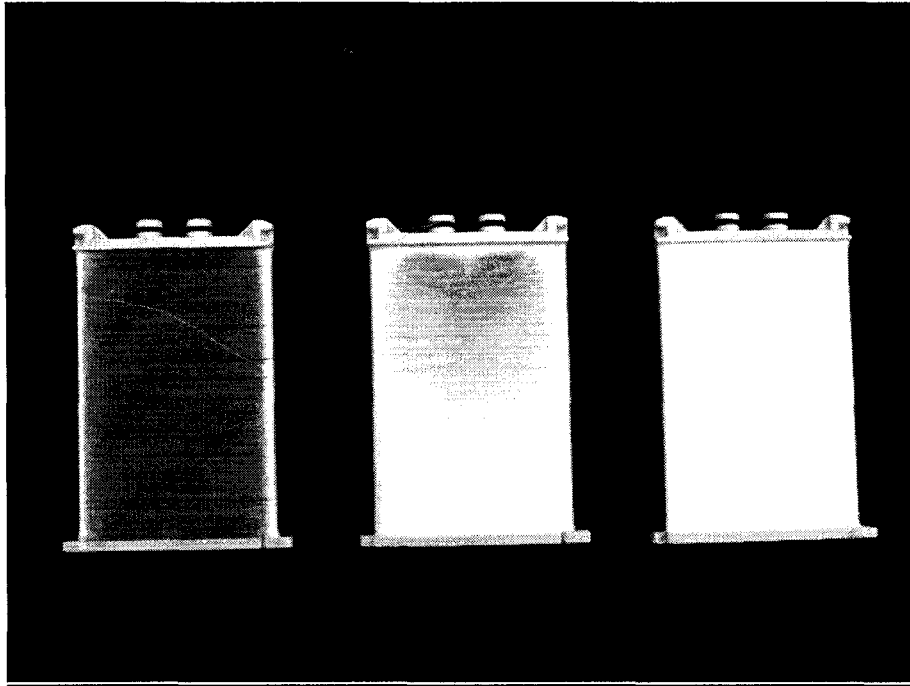


Figure 5-14 : Cake layer formation of cassette filter

CHAPITRE 6 : GENERAL DISCUSSION AND CONCLUSIONS

The general objectives of this project were to investigate the impact of seasonal water quality variations of two natural waters on their respective fouling potentials and to evaluate the reduction of this fouling potential by automatic backwash filters at a bench-scale.

6.1 Membrane clean water permeability

Due to the small size of the active membrane area used in test cell experiments (13.4 cm^2), the heterogeneous nature of membrane sheets can cause variation within the clean water permeability data obtained from the membrane coupons. It has been reported that preconditioning produces more homogenous permeabilities in membranes with values closer to those quoted by manufacturer (Jezowska et al., 2006). Therefore the coupons used in the MFI-UF measurement tests here underwent preconditioning and were rejected if they failed to meet established permeability criteria in order to minimize these variations.

Even with this conditioning, many coupons were rejected due to excessively low initial permeability, and of those coupons retained, the mean membrane permeability $1.74 \times 10^{-7} \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1} / \text{kPa}$ ($62.71 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) was lower than the permeability announced by the manufacturer $1.94 \times 10^{-7} \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1} / \text{kPa}$ ($70 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$). The phenomenon of flat sheet membrane samples being characterized by lower membrane permeabilities than those specified by the manufacturer has been observed in other work. It is believed that this is due to the fact that specifications given by manufacturers typically apply to membrane modules and not small areas of membrane sheets (Jezowska et al., 2006).

The small amount of information that is provided by the membrane manufacturers is a challenge to developing a standardized approach to laboratory membrane fouling tests. In the present study, the variations noted were independent of batch effects as all samples came from the same manufacturing batch. Future works should propose guidelines outlining the degree of variation between coupon c that can CWP considered acceptable for laboratory testing of membrane fouling.

6.2 Pre-filtration with laboratory filters – a cautionary tale

In the past, researchers have used laboratory filters to examine the role of particle / colloid size on fouling potential as well as to model the performance of membrane pre-treatments (Kumar et al., 2006; DiGiano et al., 2000). This practice is based on the presumption that the filters used will remove the majority of particles larger than the pore size announced and allow the majority of particles smaller than the pore size to pass through. As the results of preliminary tests conducted here with laboratory polypropylene filters show, it is not always possible to assume with certainty that laboratory disc filters will perform the way proclaimed, and verifications should be made whenever possible when experiments are performed with laboratory filter products

6.3 Seasonal water quality and fouling potential analysis

As presented in the article (chapter 4), results of the analyses carried out on the seasonal water quality and fouling potential variations suggest that traditional water quality parameters cannot be used to determine the presence or concentration of foulants amongst different water sources. It was also found that the variable that played the most important role in the general model developed to explain fouling potential variability was

the water source. However, the models that were developed for single water sources were highly successful at explaining the variability of fouling potential as a function of the water quality parameters that were measured. This implies that the influence of water quality on the MFI-UF and therefore on fouling potential is dependent on the water source.

General correlations between fouling potential and the measured water quality parameters (turbidity, TOC, UV₂₅₄, particle counts, Ca, Mg and Fe concentrations, etc) could not be made. This had been previously noted in the literature (Yiantsios et al., 2005; Howe et Clark, 2002b; Heijman et al., 2005). However, in the specific cases of the two water sources studied, certain parameters displayed strong correlations with fouling potential. In the case of Barron Lake there was a strong correlation between TOC and fouling potential suggesting that influential foulant(s) (possibly a specific fraction of the NOM) varied in proportion with the total organic carbon. In the same manner, turbidity was strongly correlated with fouling potential both for the Des Prairies river and Barron lake separately, suggesting that a specific fraction of turbidity which likely played an important role in fouling varied proportionally with turbidity in both cases.

The characterizations of Barron Lake and the Des Prairies River over an eight month period facilitated the development of profiles for the two water sources thus providing a context to better interpret variations in fouling potential. The characterizations also provided information on the role that weather events might have on fouling potential. The spring turnover and snowmelt were identified as a challenging period in terms of fouling potential increase for the Des Prairies River, while it was a precipitation event during the summer and another unidentified event at the end of October (possibly an algal bloom) that caused an increase in fouling potential at Barron Lake.

Future works should aim at identifying and developing quantification methods that specifically target possible foulants (colloidal matter, protein and polysaccharide content, etc) in feed waters. In parallel a standard procedure could be developed to profile sources in terms of surrogate fouling measures (e.g. turbidity and TOC at Barron Lake) which could then be easily used to follow the evolution of fouling potential throughout the year. This would allow existing facilities to put in place more aggressive fouling management practices during the periods that are known to be problematic. For future installations, the profiling of a source could be useful for effective planning of piloting schedules to ensure pilots are in place when the most challenging fouling potential conditions occur.

6.4 Membrane Pre-treatment evaluation

As mentioned previously, the bench-scale pre-filtration protocol developed here used samples of screens and thread filters employed in full scale installations. It is believed that this protocol better estimated the performance of automatic backwash filter technology than traditional laboratory filtration could. This is because the latter is typically performed with filters made of different materials than automatic backwash screens / thread filters are made of.

Particle counts of the filtrates obtained through the aforementioned protocol showed that the pre-filters removal of particles larger than the announced porosity was not absolute. However some of the filters did provide a certain degree of protection against larger particles, for example, there was a 74% removal of particles with diameters greater than 2.25 μm by the 2 μm pre-filter. This removal had little to no effect on the reduction of the MFI-UF; in fact none of the pre-filtration had a significant effect on the fouling potential. It appears thus that degree to which particles larger than 2 μm are removed plays a negligible role on fouling potential reduction. Caution should be exercised when

selecting a pre-treatment which removes particles in the aforementioned size range as the announced porosity is not a good parameter for describing the performance of the pre-filter in terms of fouling potential reduction.

A cake layer was expected to form on the surface of the automatic backwash filter over the duration of a filtration run before the cleaning cycle is activated. It was therefore considered that this cake layer might affect the particle removal of the filter. The formation of a cake layer on a 2 μm cassette filter at constant flowrate did not however improve the removal of particles of diameter greater than 2.25 μm . There was actually a small increase in the particle counts towards the end of the filtration run probably due to the increasing shear forces that developed as the run progressed. The slight decrease in turbidity suggested however that there was a small increase in the removal of particles smaller than the detection limits of the DPA 4100 particle counter. The removal of the smaller particles by the cake layer was insufficient to significantly alter the fouling potential of the filtrate samples.

It should be noted that while the removal of large particles provided no advantages in terms of fouling potential reduction for the membrane flatsheet itself, it is likely that the removal of larger particles could prevent the plugging of feed spacers within spiral wound membrane modules. It seems unlikely however that the use of coarse pre-filtration ($>30 \mu\text{m}$) would be useful for this protection since the concentrations of particles larger than 30 μm were typically very low compared with the concentrations of other particle sizes for the two sources studied.

The results presented here support findings published that suggest colloids are responsible for fouling (Chellam et al., 1997; Jarusutthirak et al., 2002; Park et al., 2006; Amy et al., 2001; Yiantsios et al., 2005). Pre-treatment for spiral wound membranes should therefore be capable of removing these small particles and colloids in order to

effectively reduce the fouling potential. Future works should focus on investigating which treatment techniques can feasibly remove sub-micron matter without overly increasing costs.

6.5 Conclusions

The main objectives of the project were attained. The investigation of the impacts of seasonal water quality variations on fouling potential resulted in the determination that traditional water quality parameters are inadequate to generally describe or quantify foulants, but that these parameters can be useful in following fouling potential when the profiles of specific water sources are known. The evaluation of the reduction of fouling potential by automatic backwash filters at a bench-scale was also successful and it was determined that the size of particle removed by this technology (2 μm and larger) does not play an important role in fouling, and therefore fouling of the membrane flatsheets would not be reduced through this pre-treatment.

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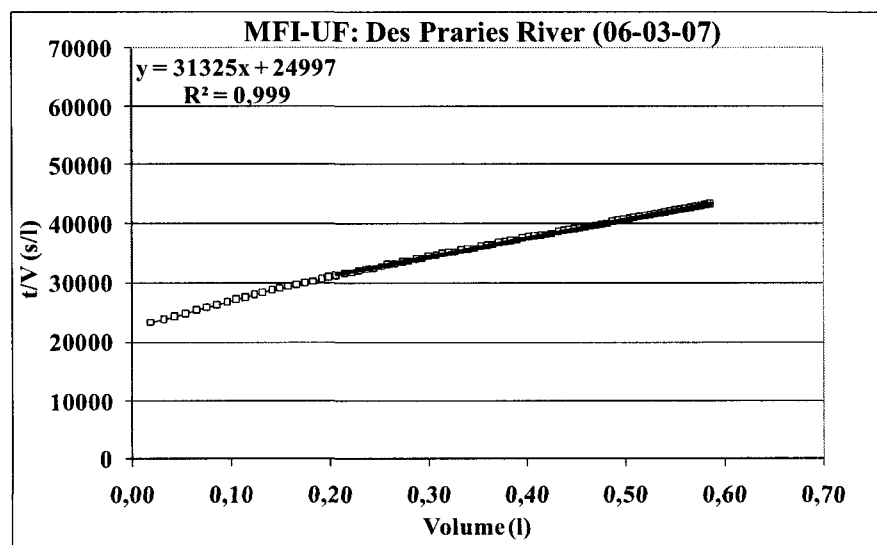
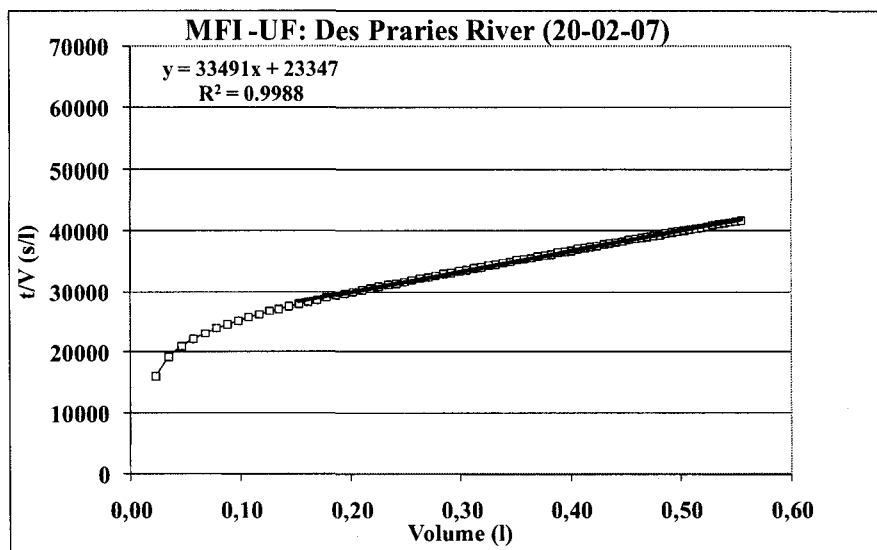
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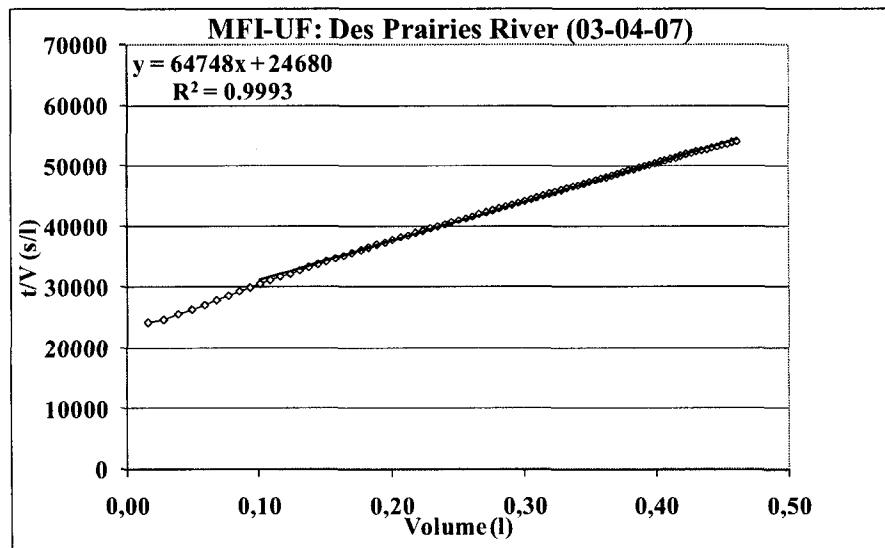
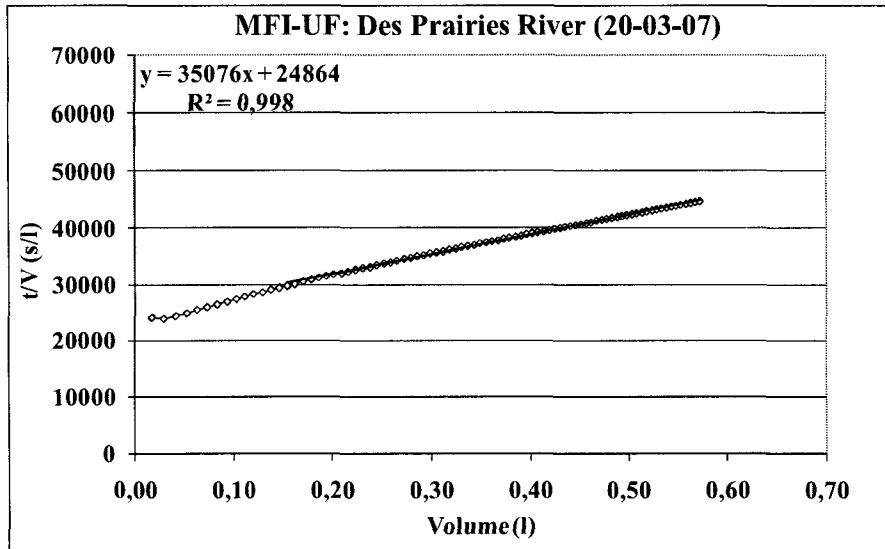
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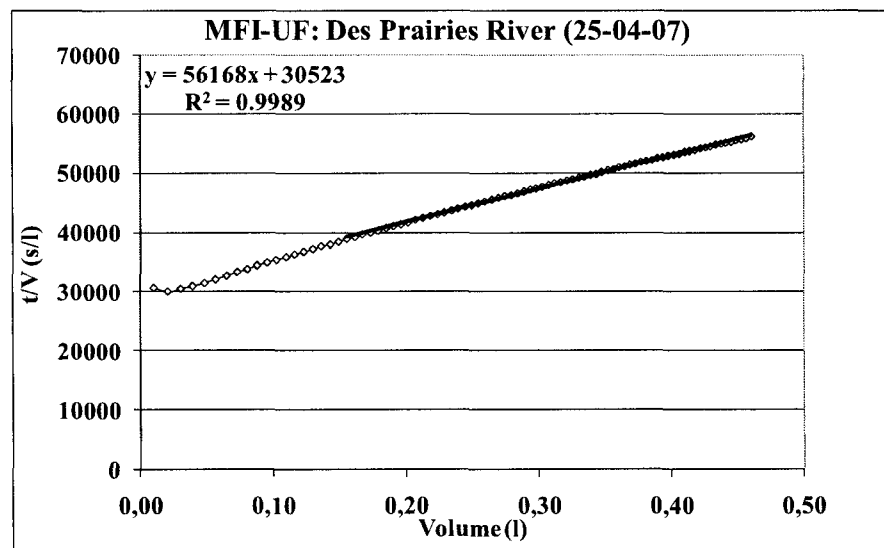
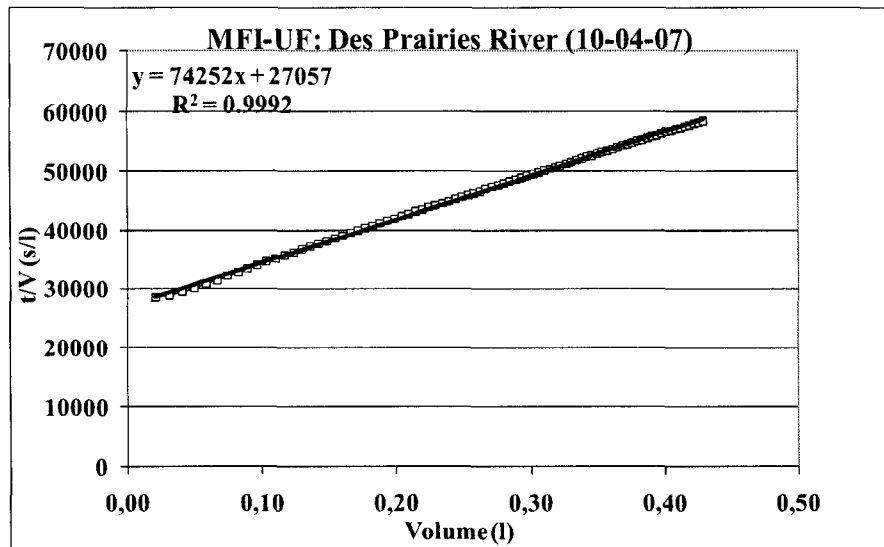
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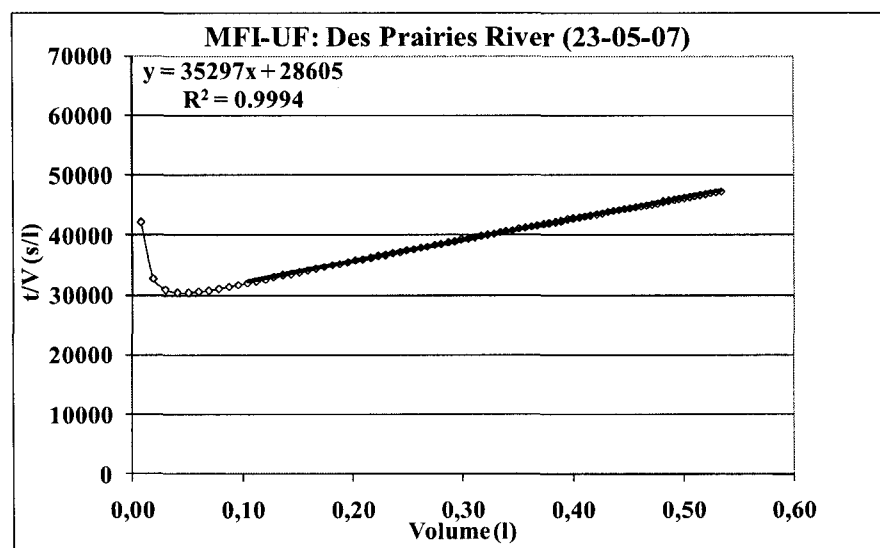
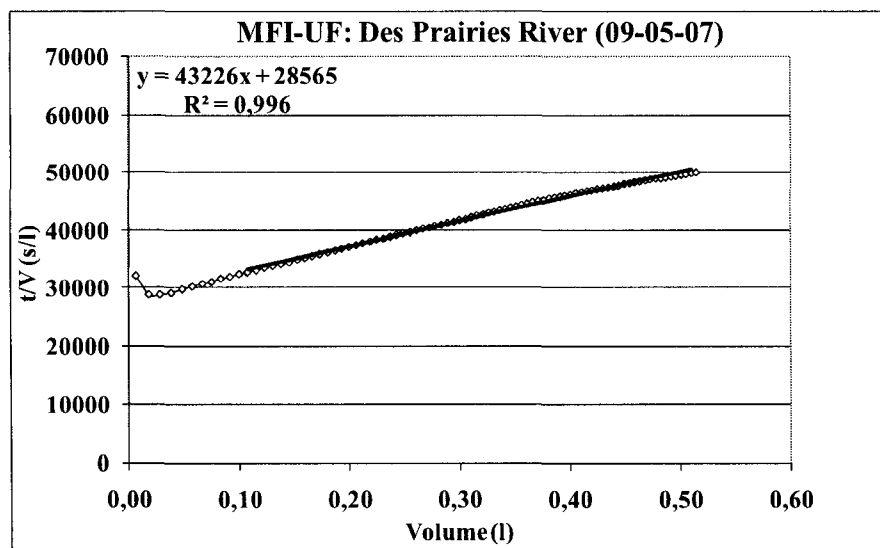
APPENDICES

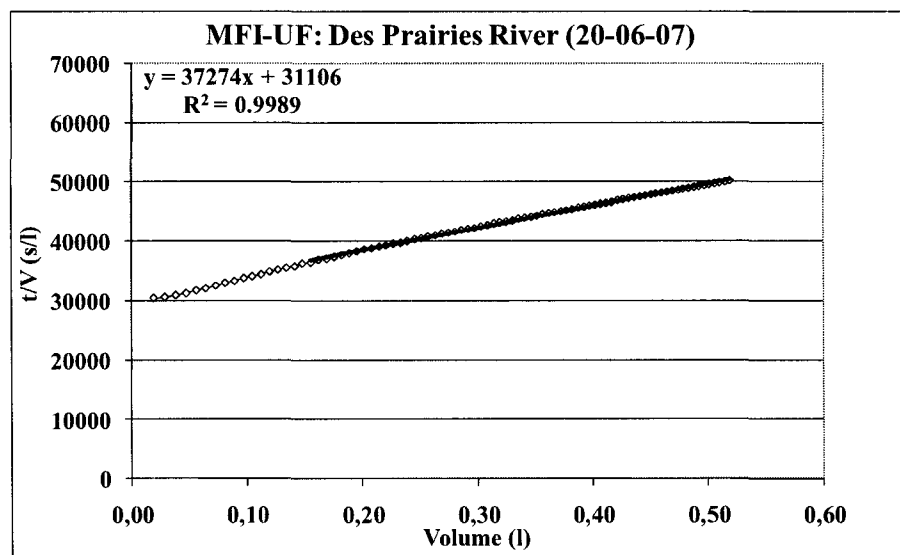
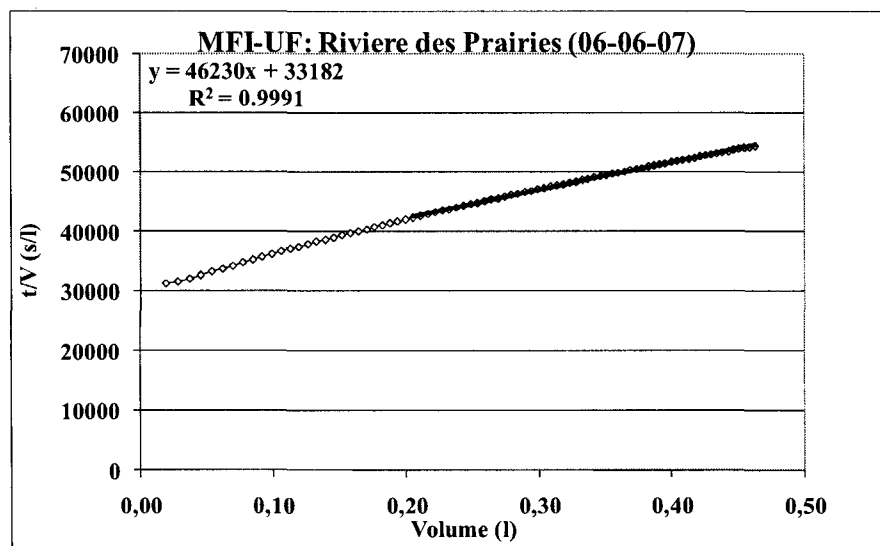
APPENDIX 1 : MFI-UF MEASUREMENTS

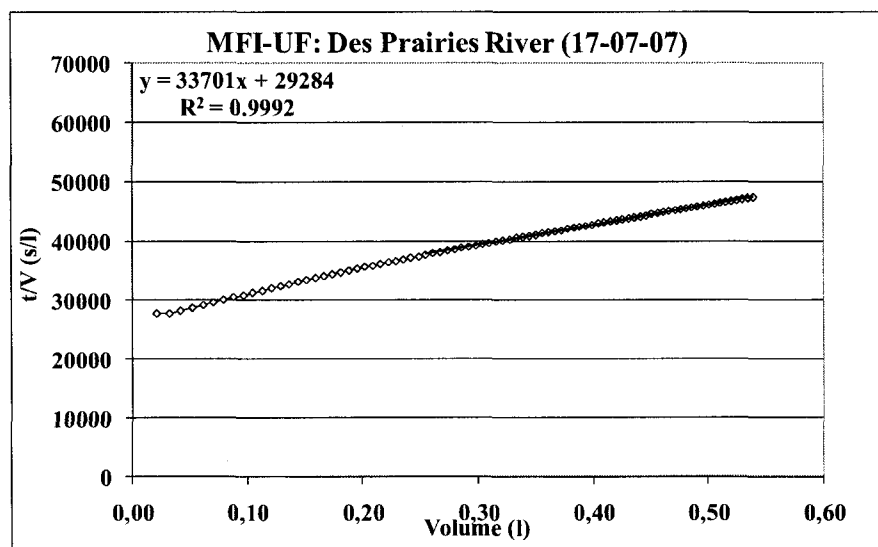
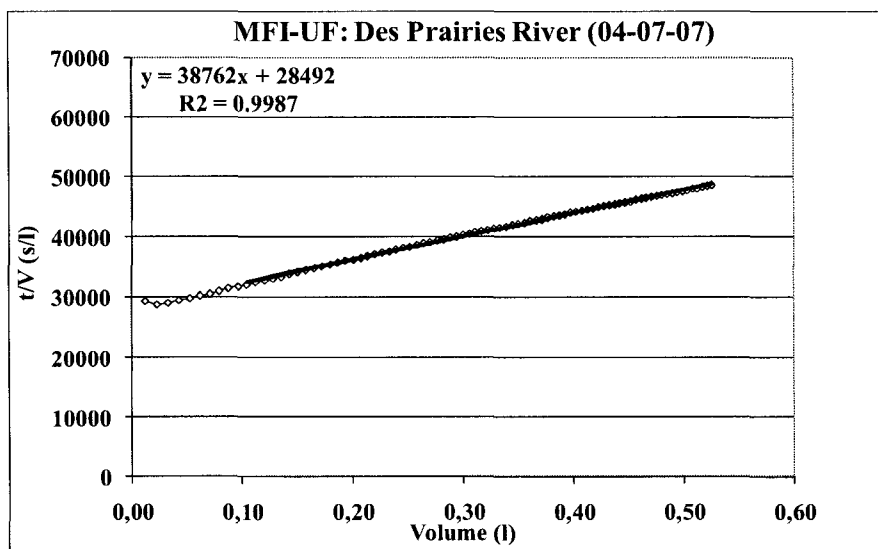


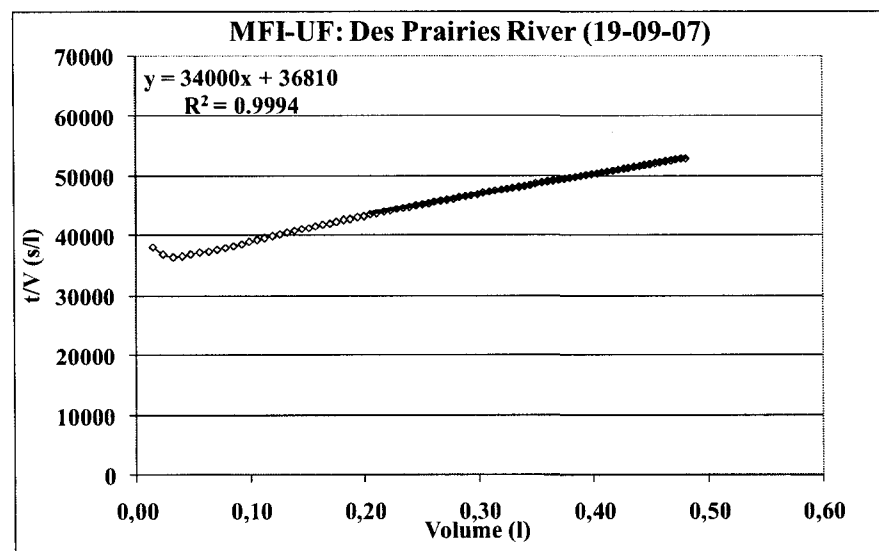
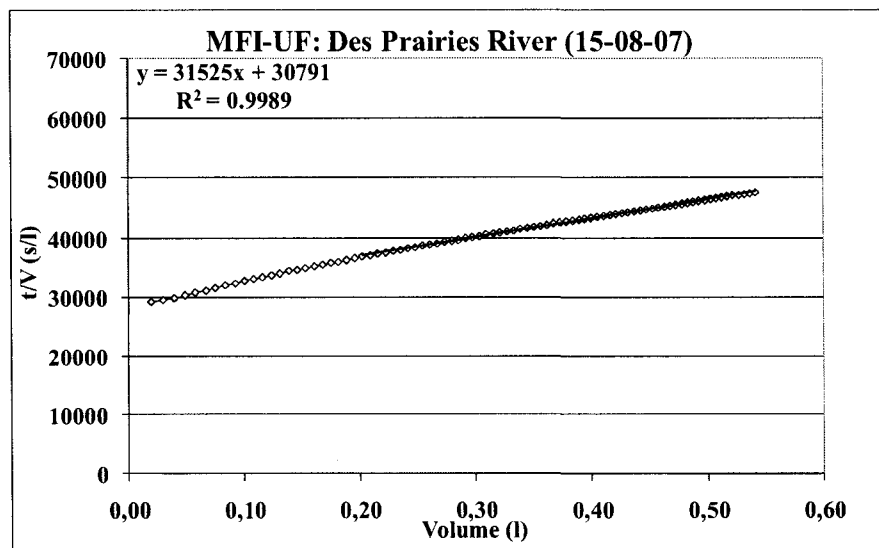


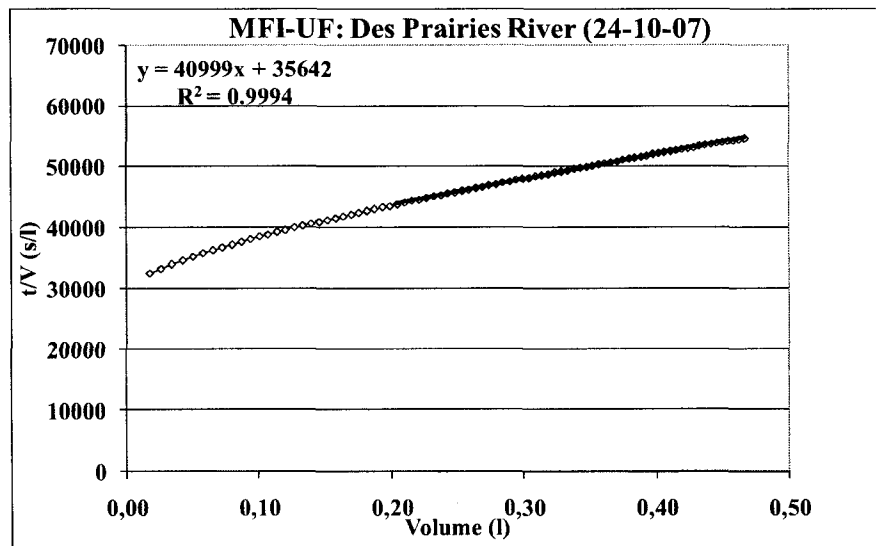
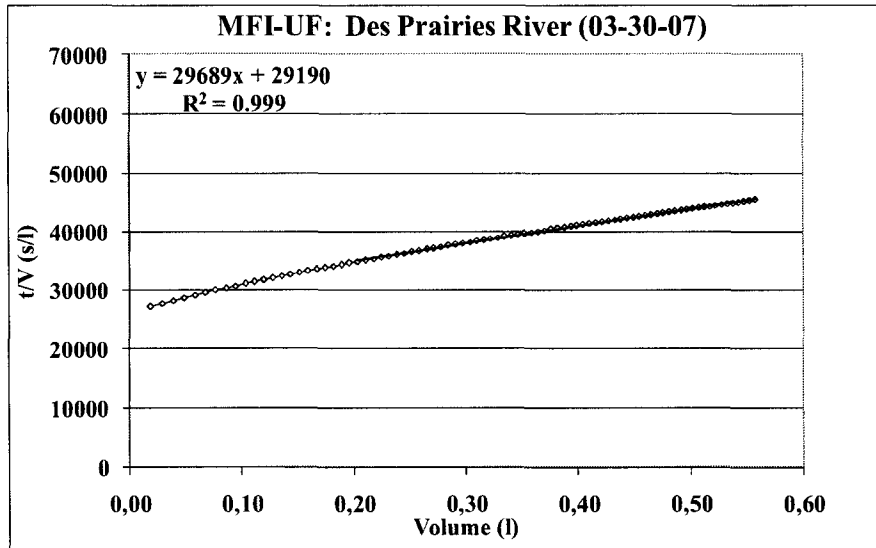


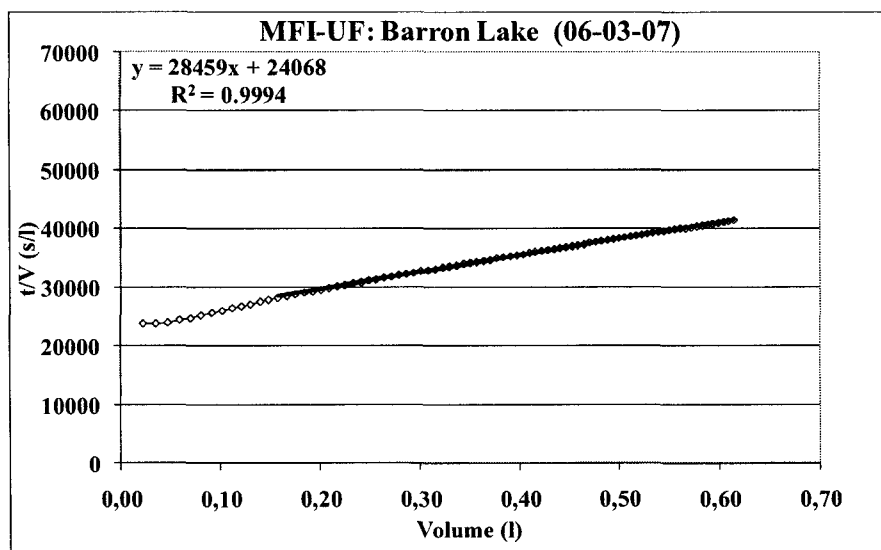
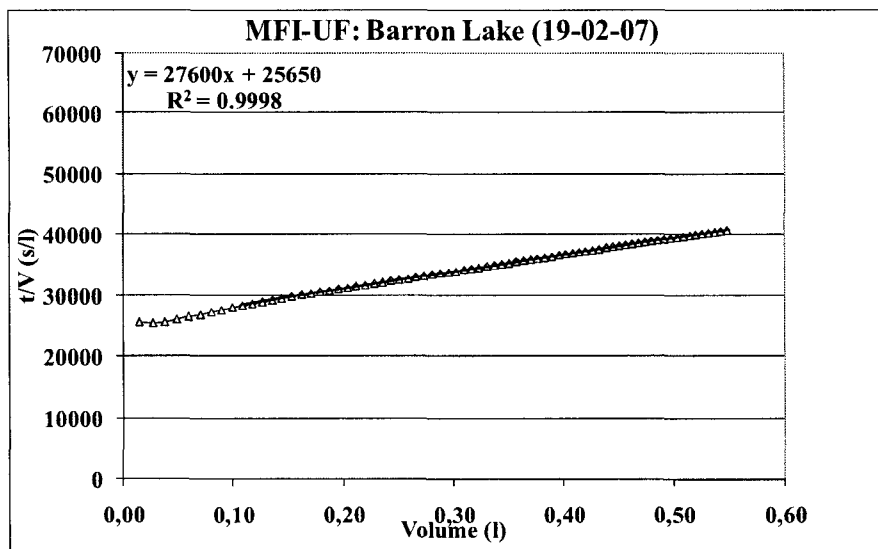


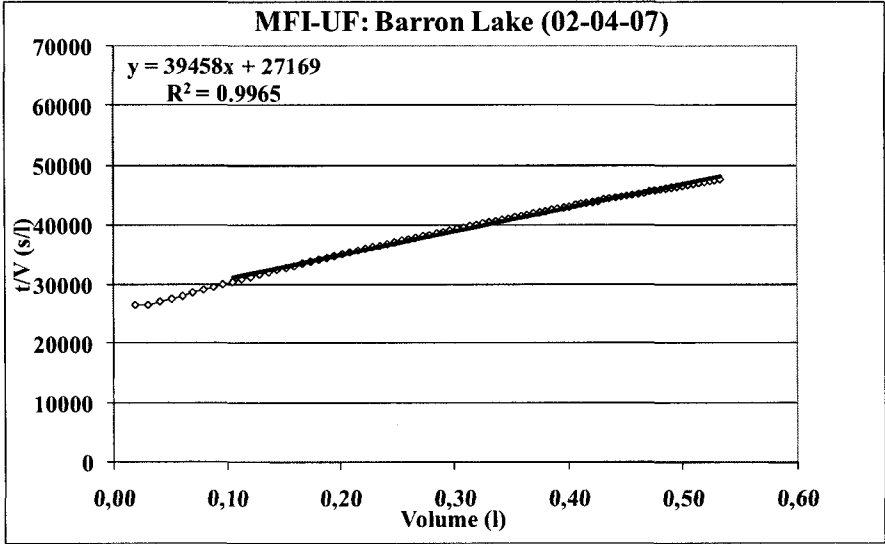
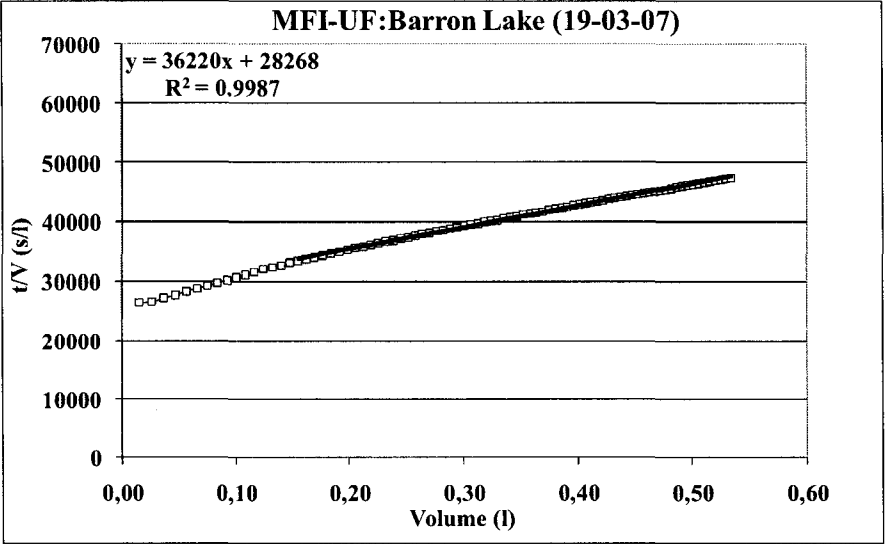


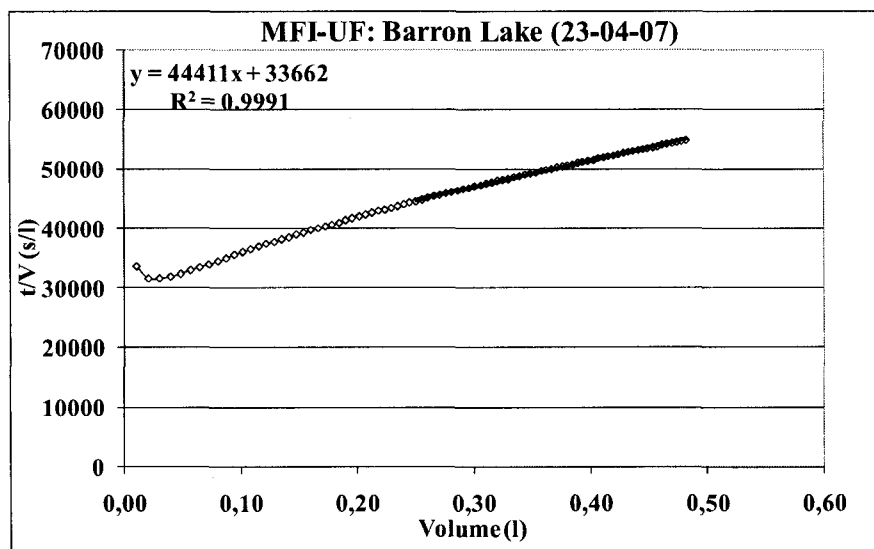
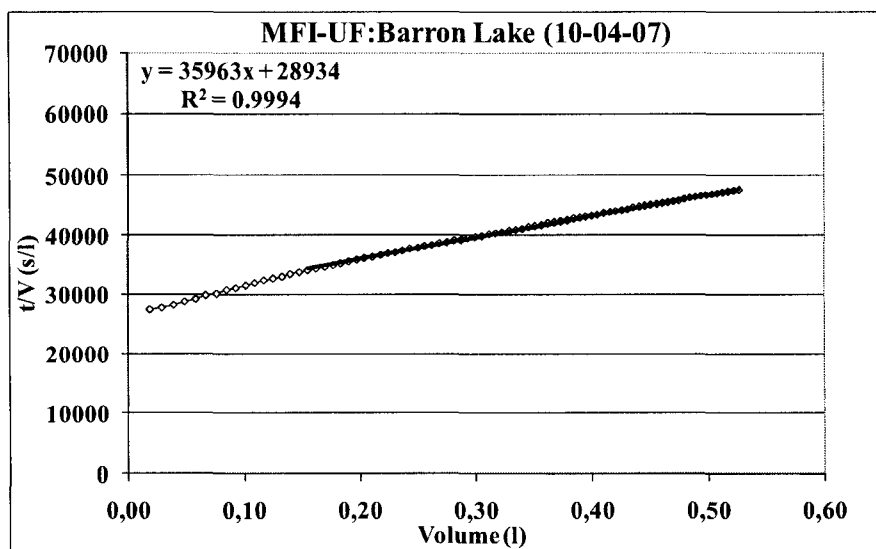


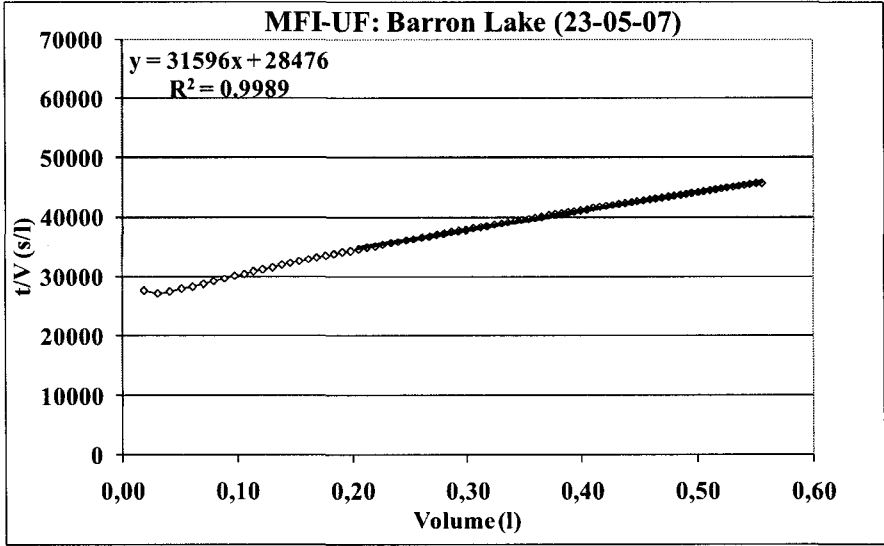
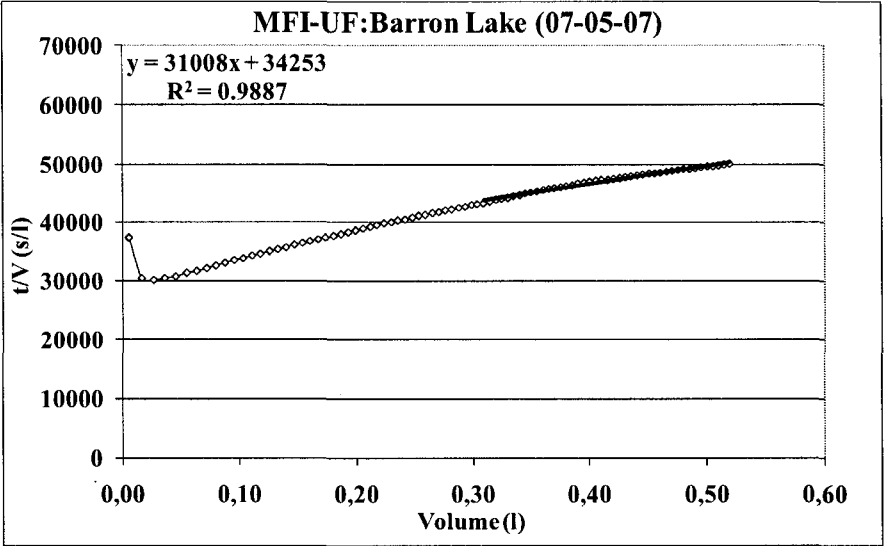


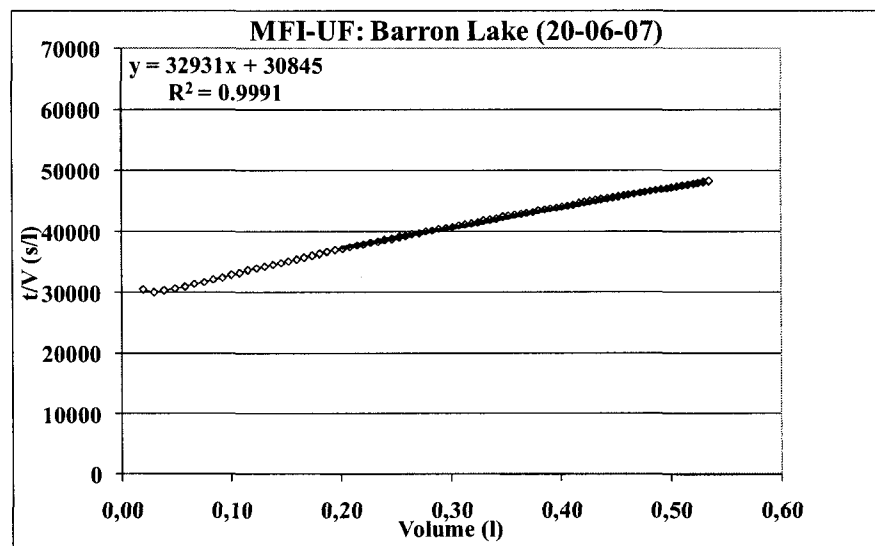
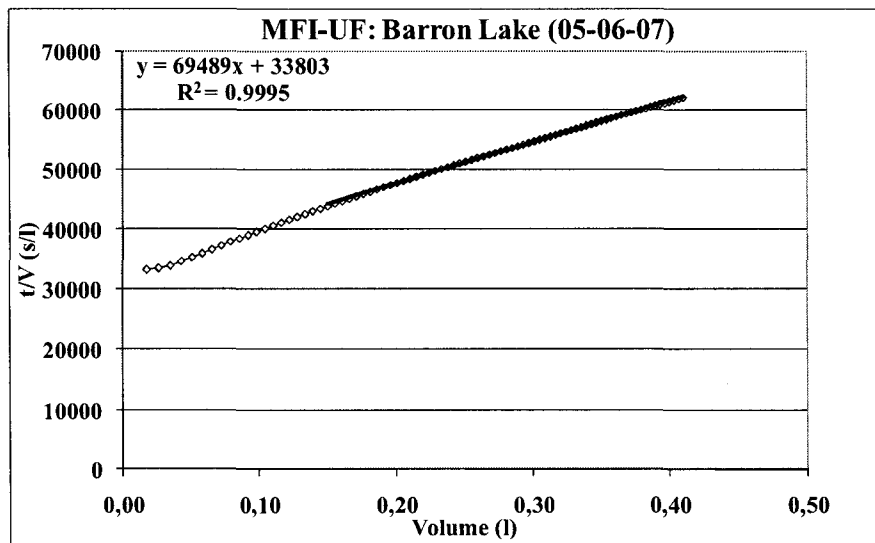


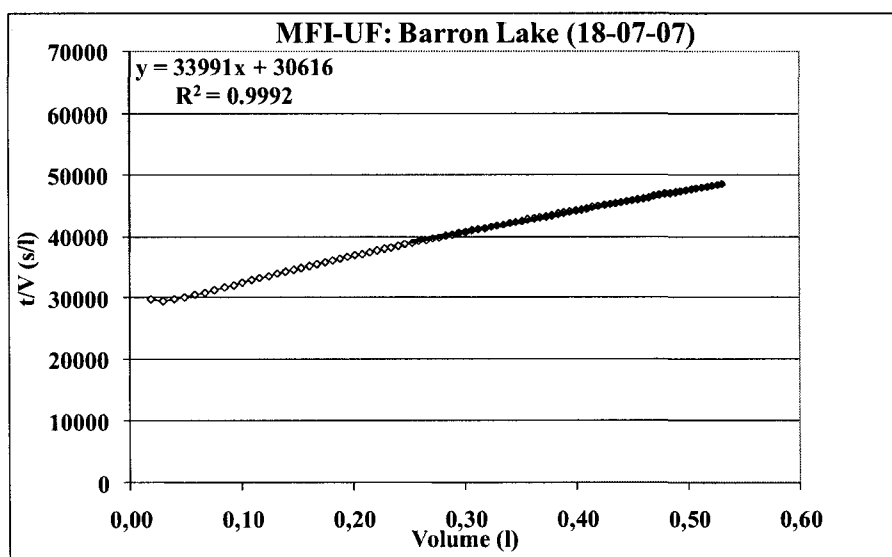
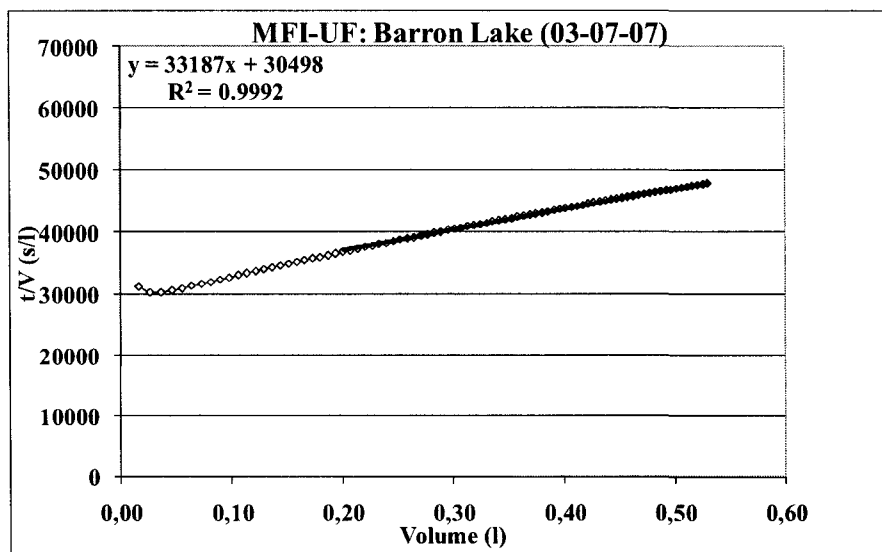


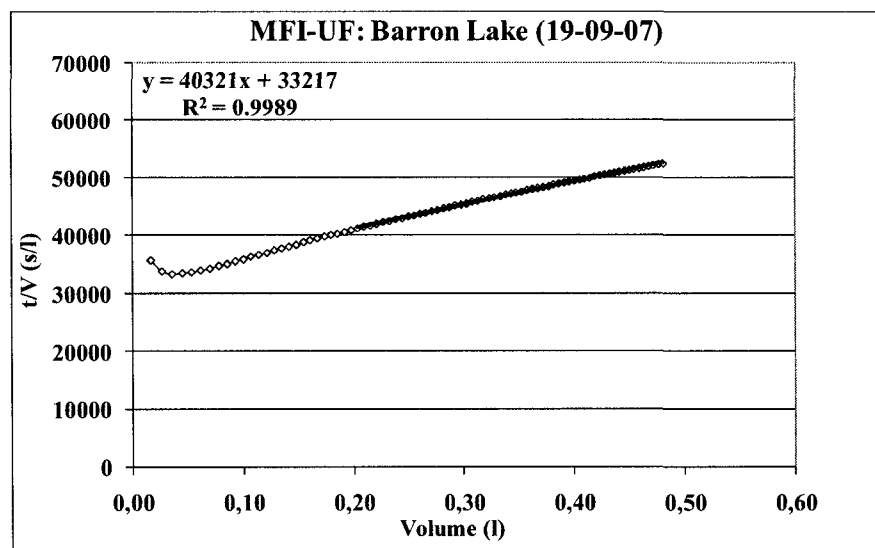
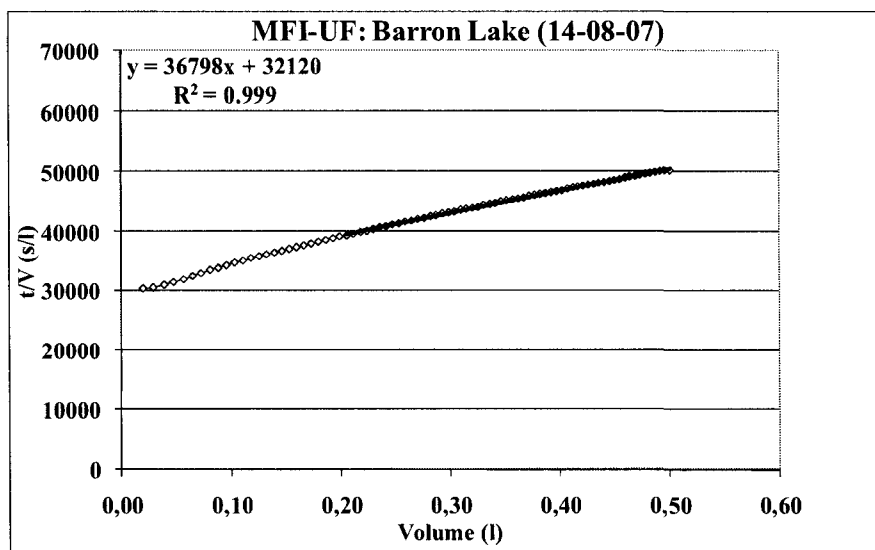


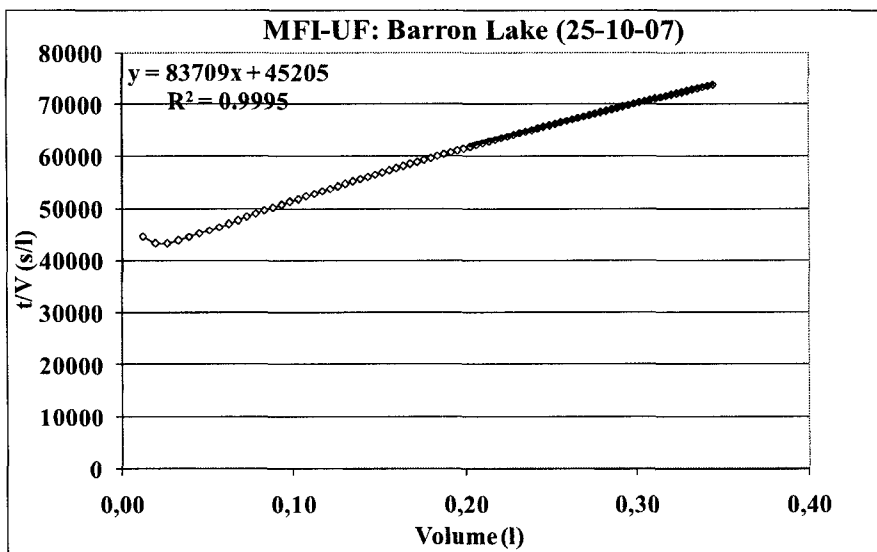
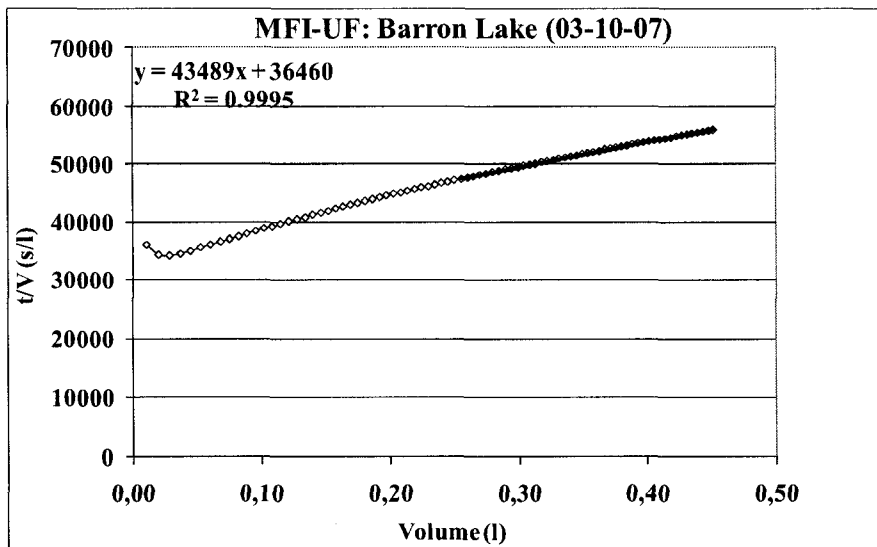


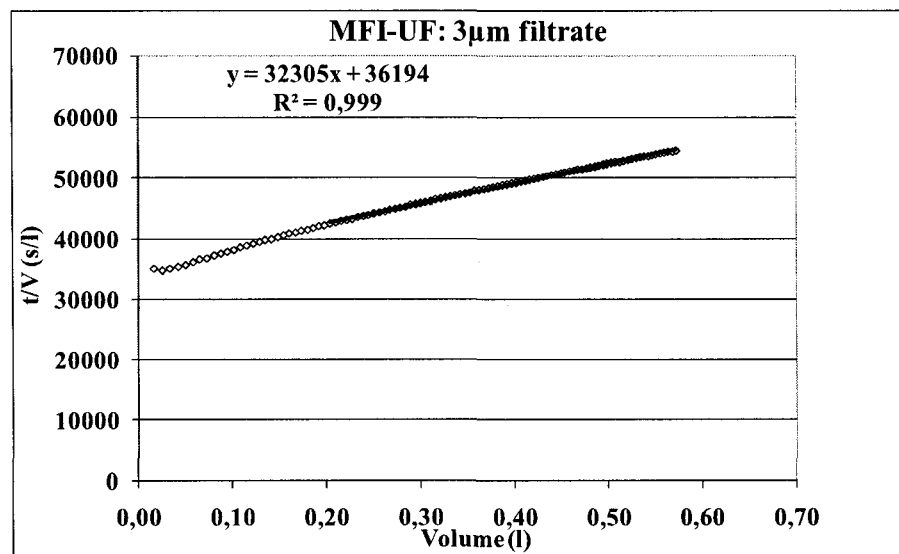
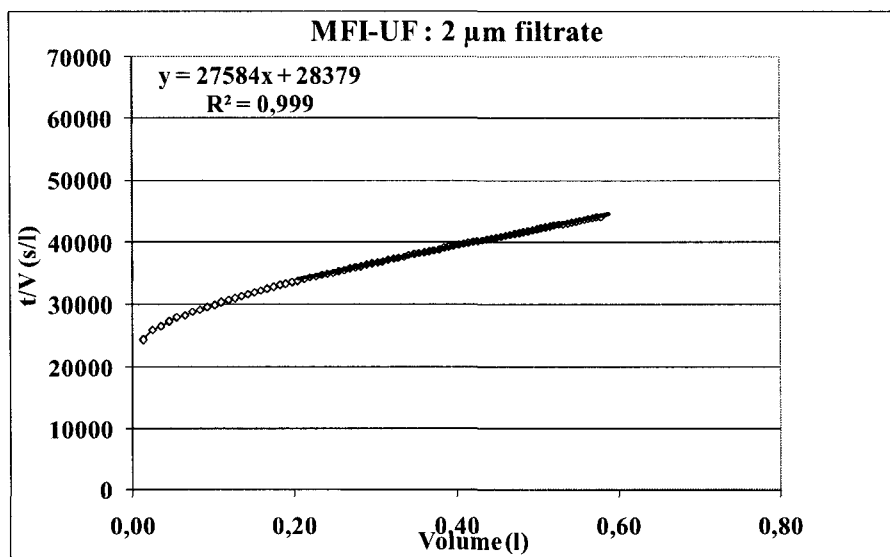


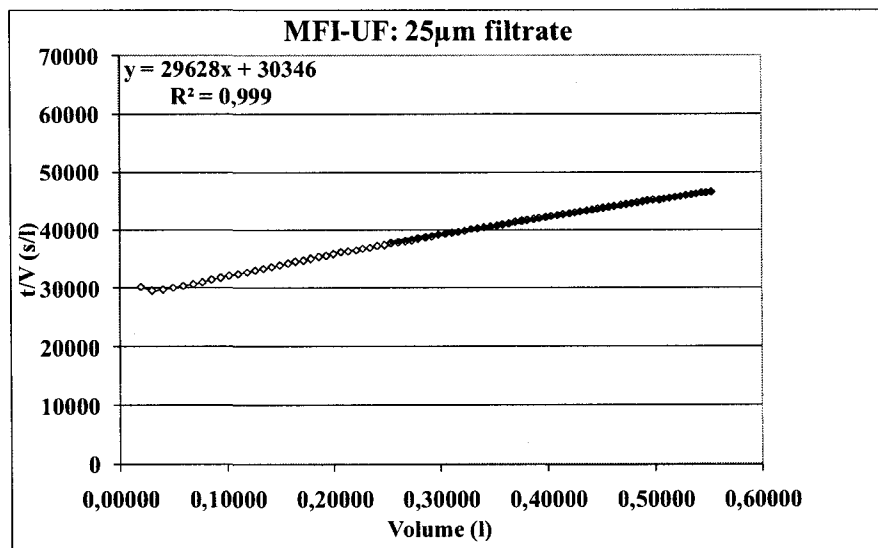
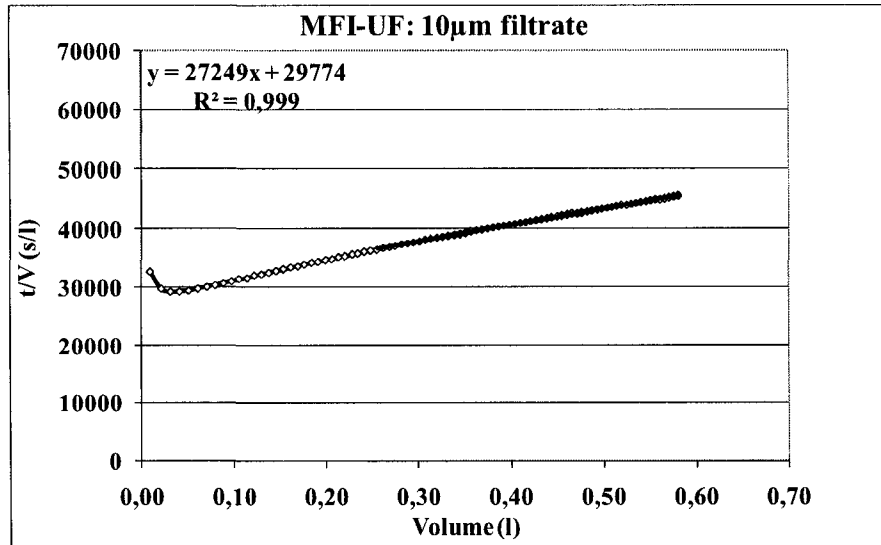


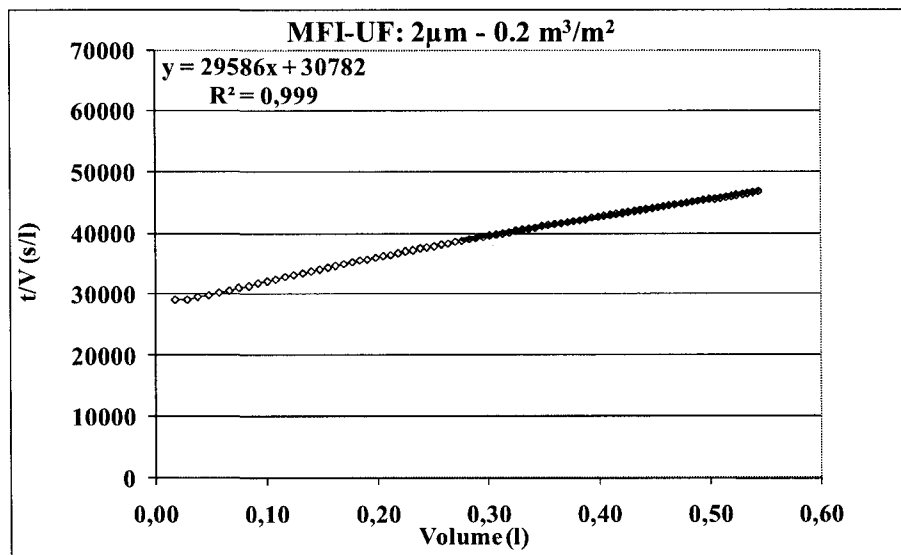
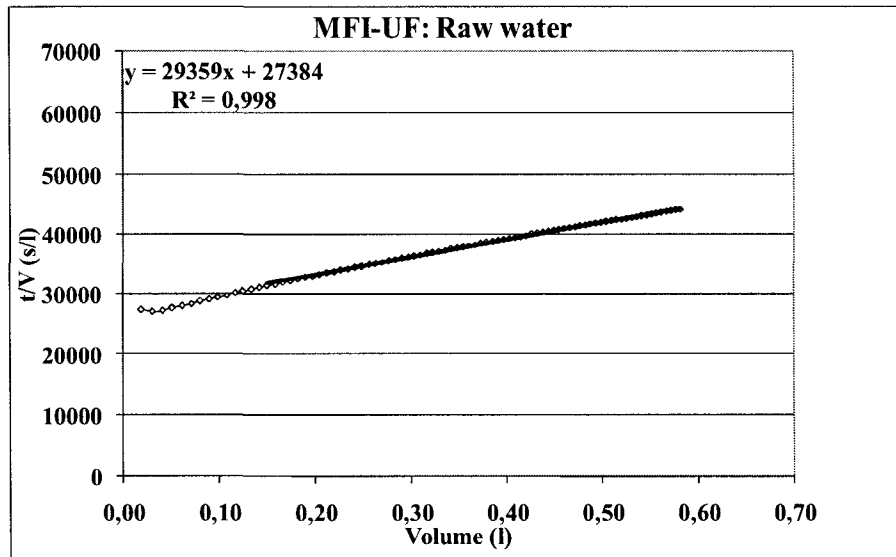


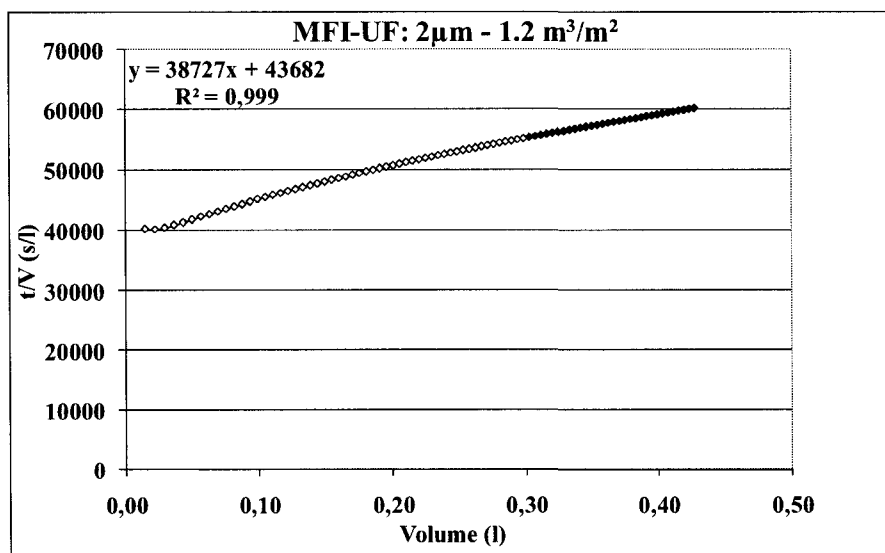
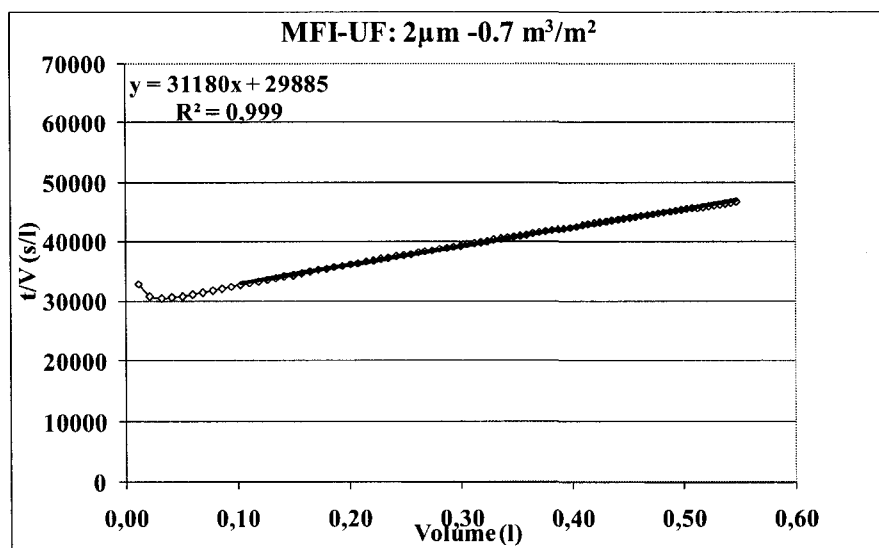


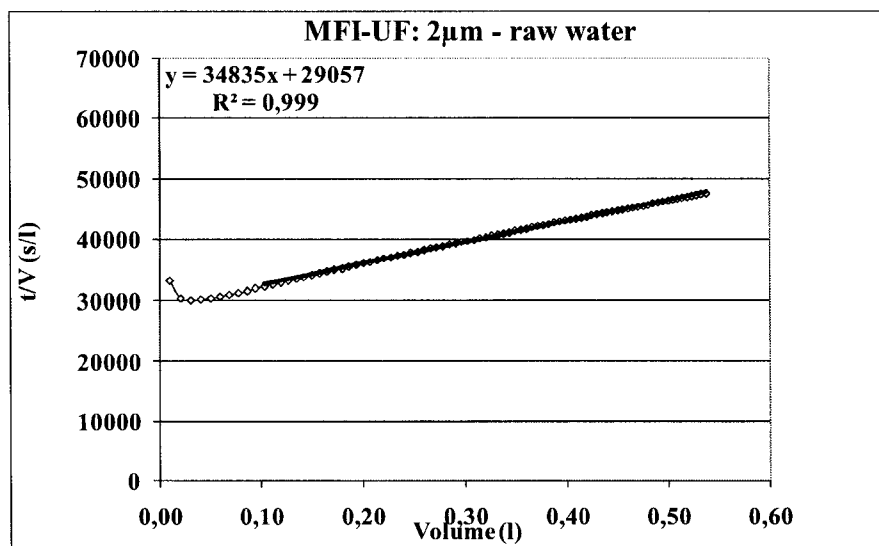


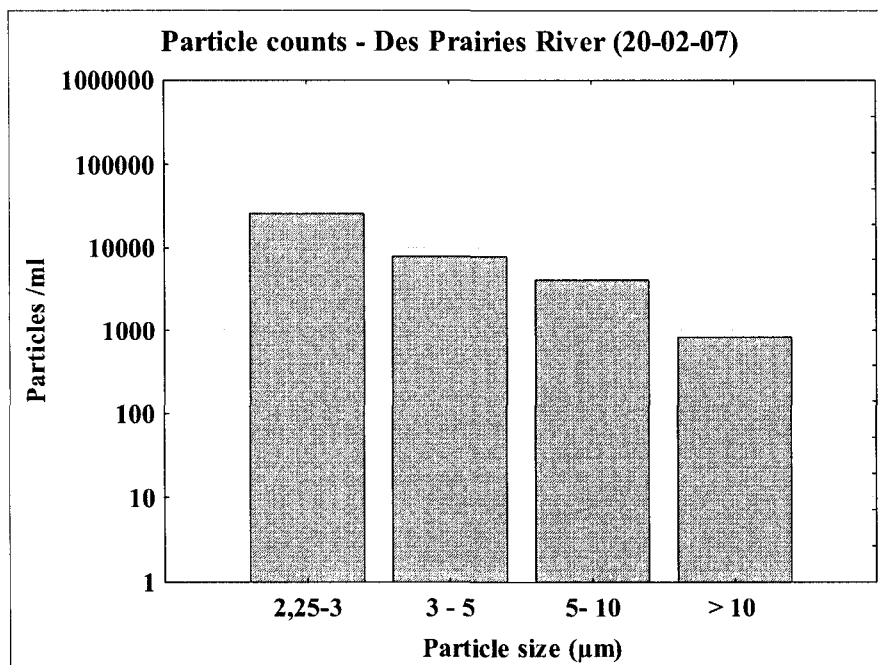


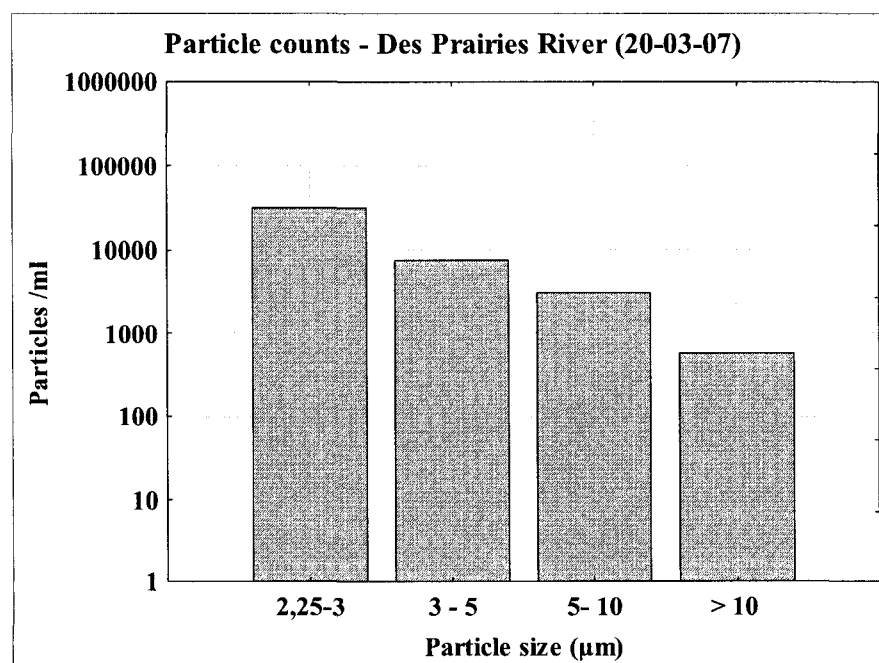
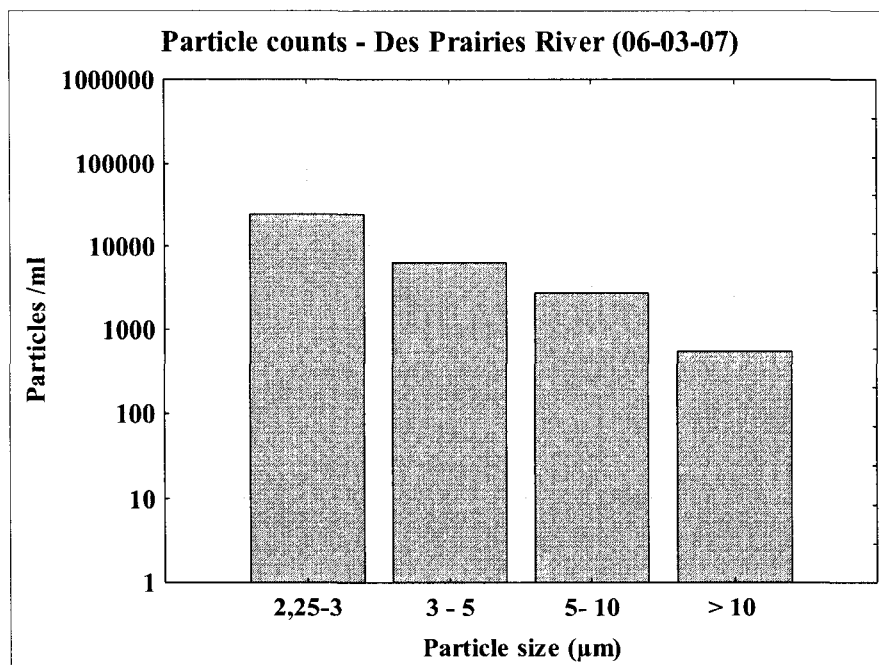


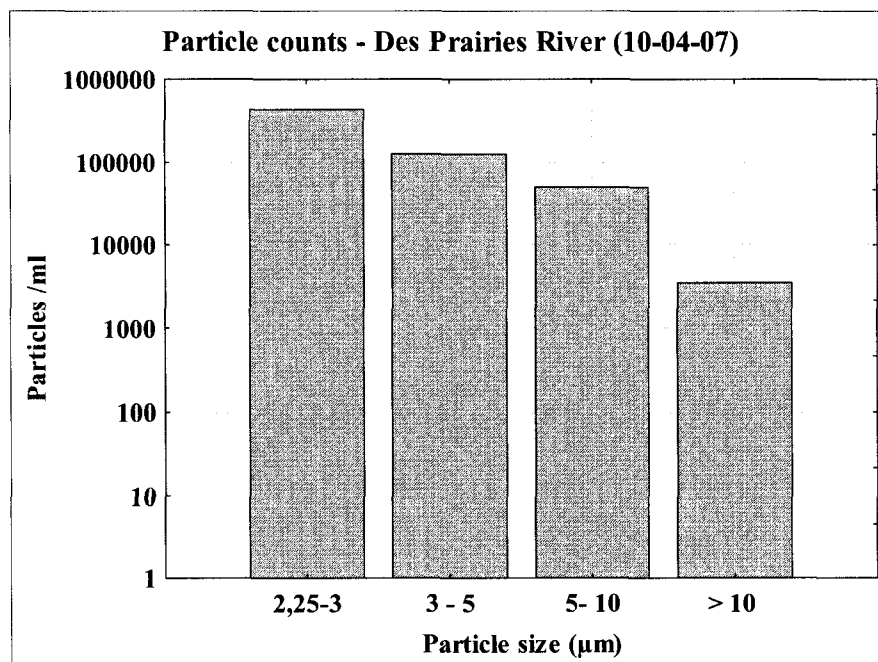
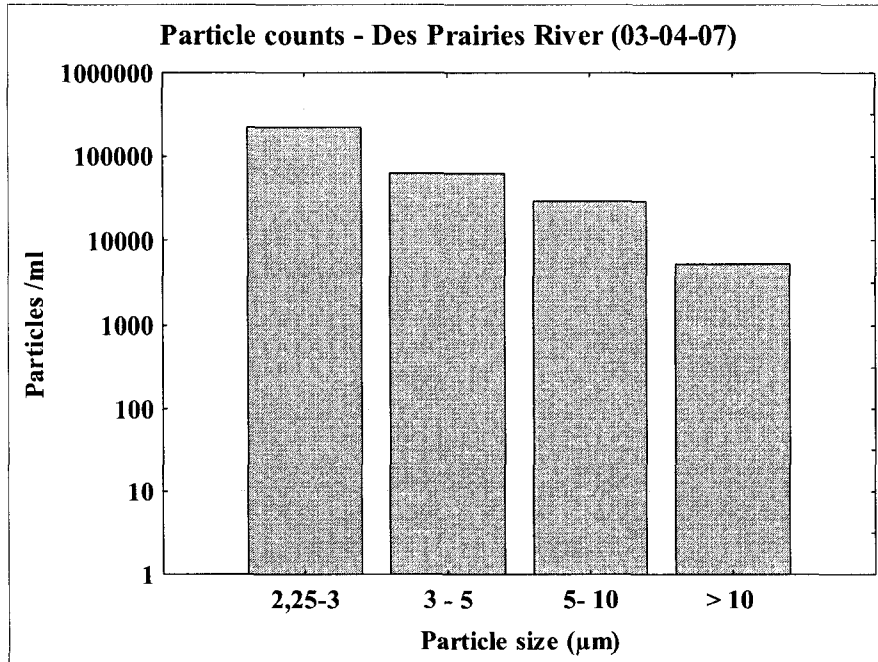


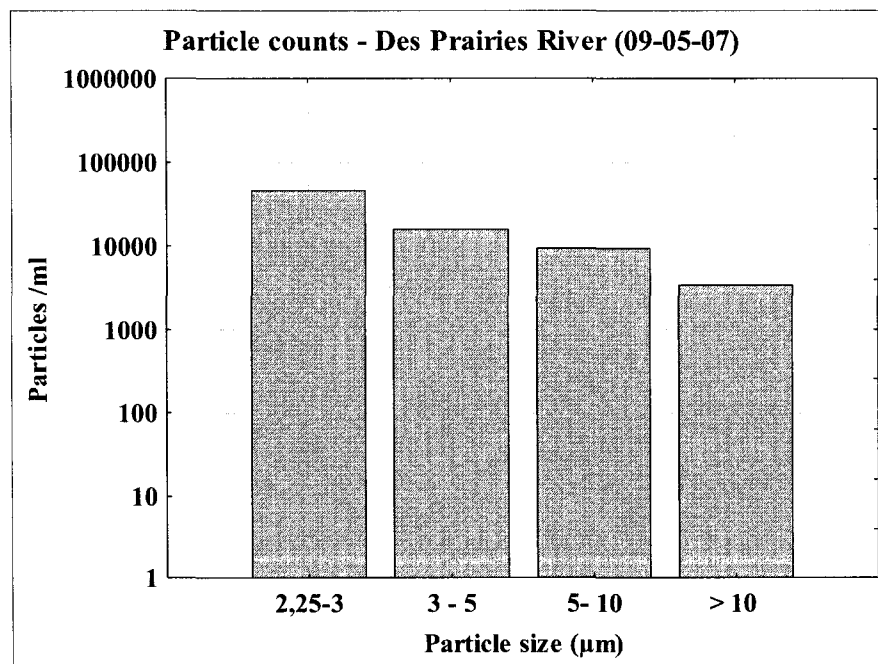
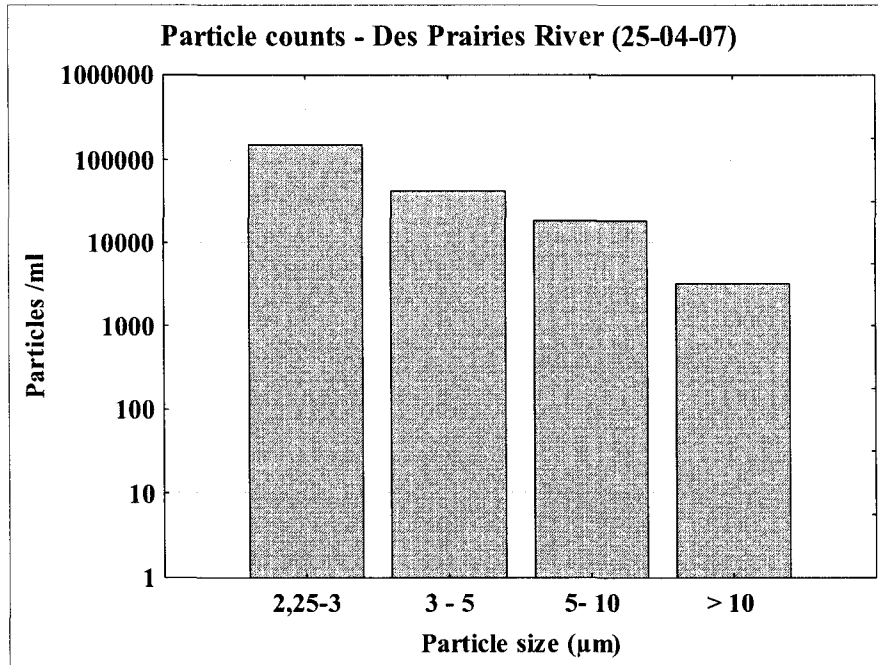


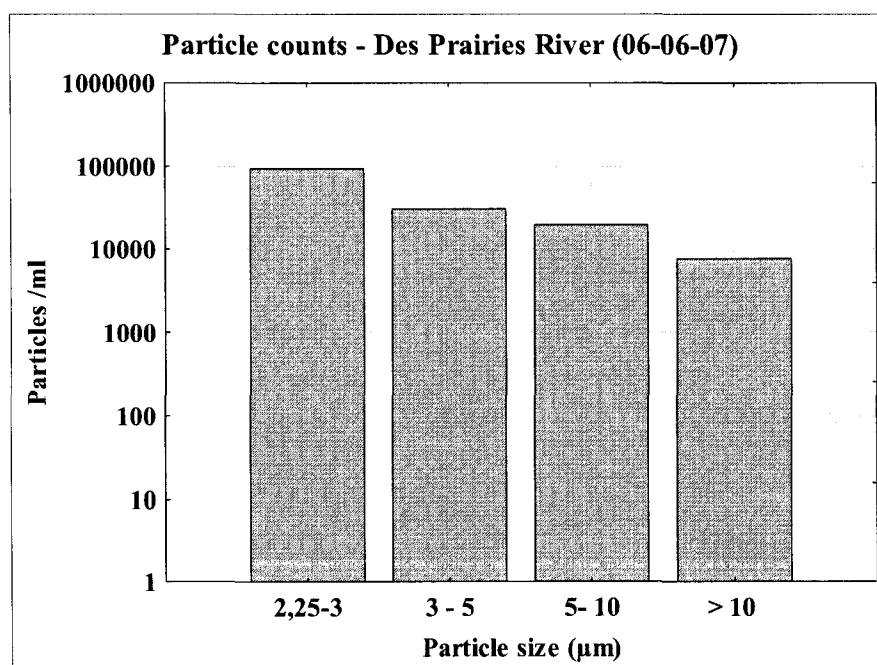
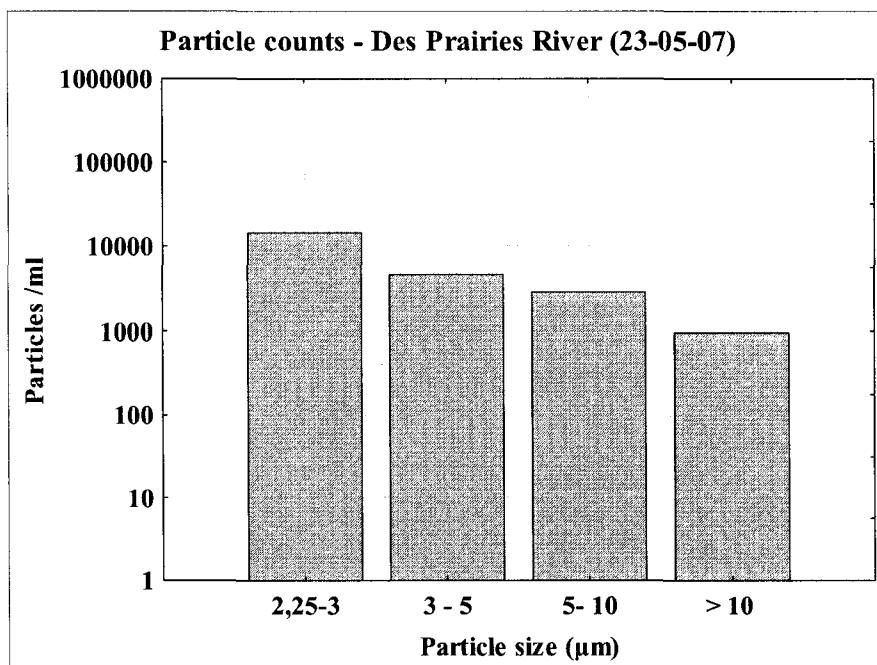


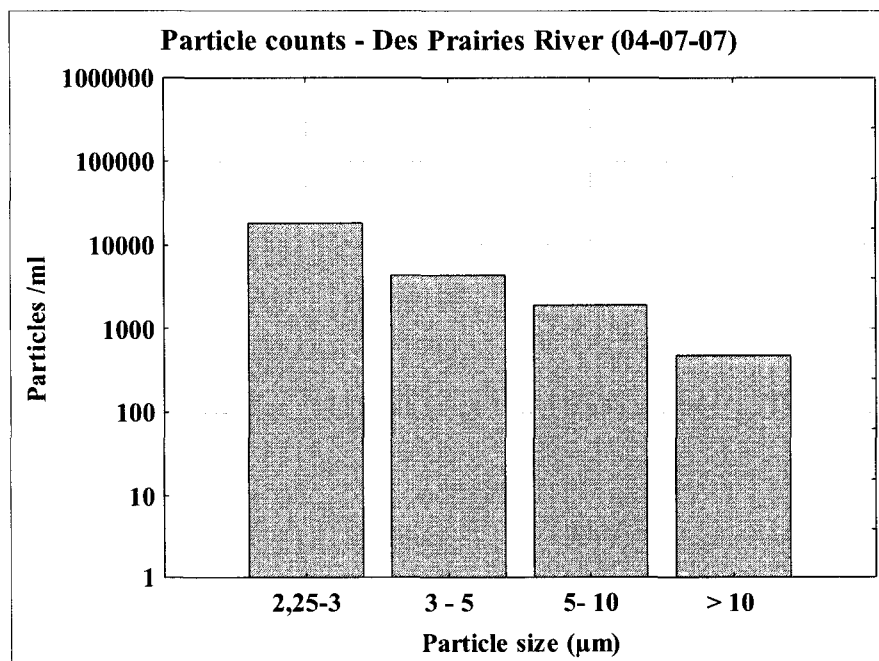
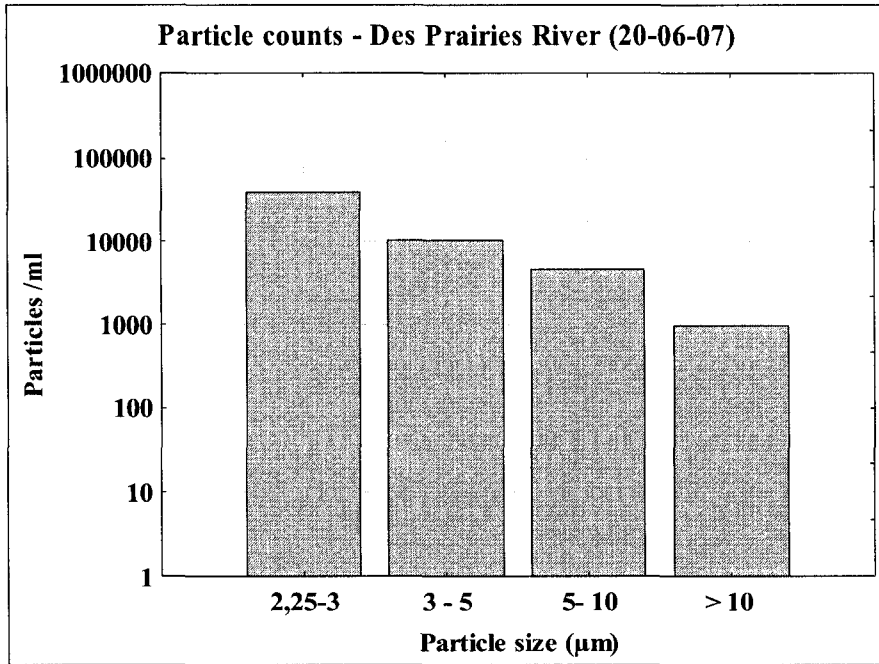
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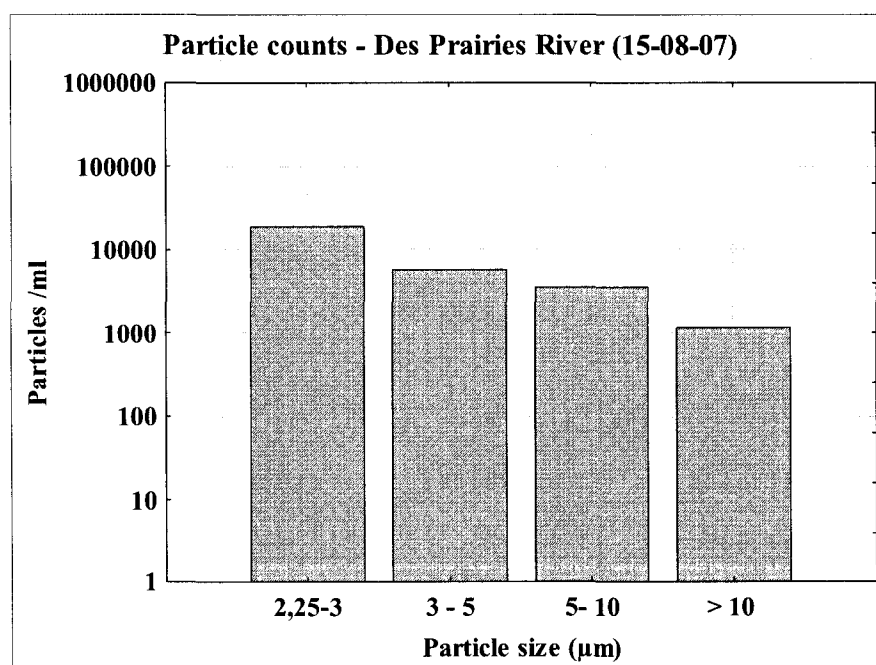
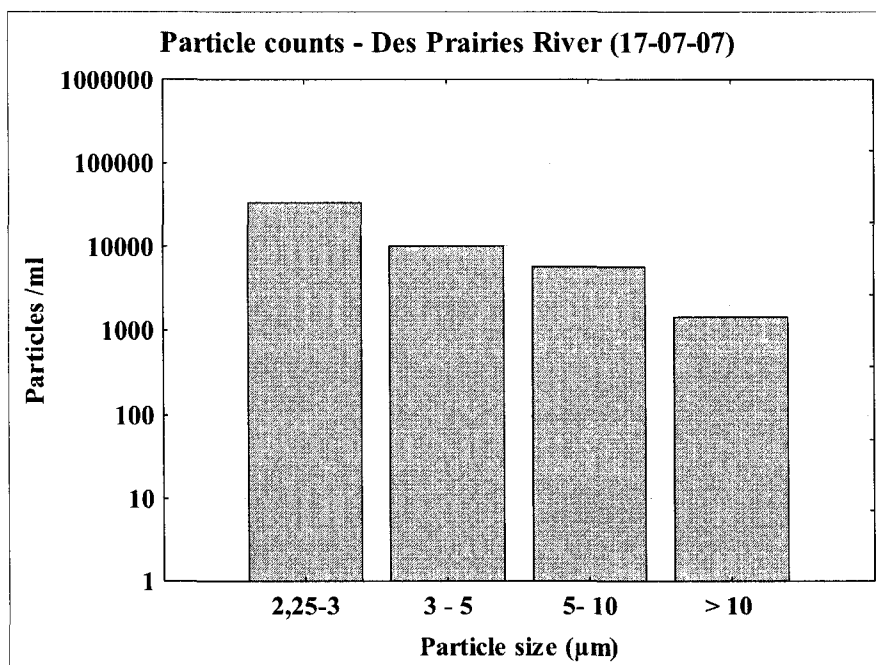


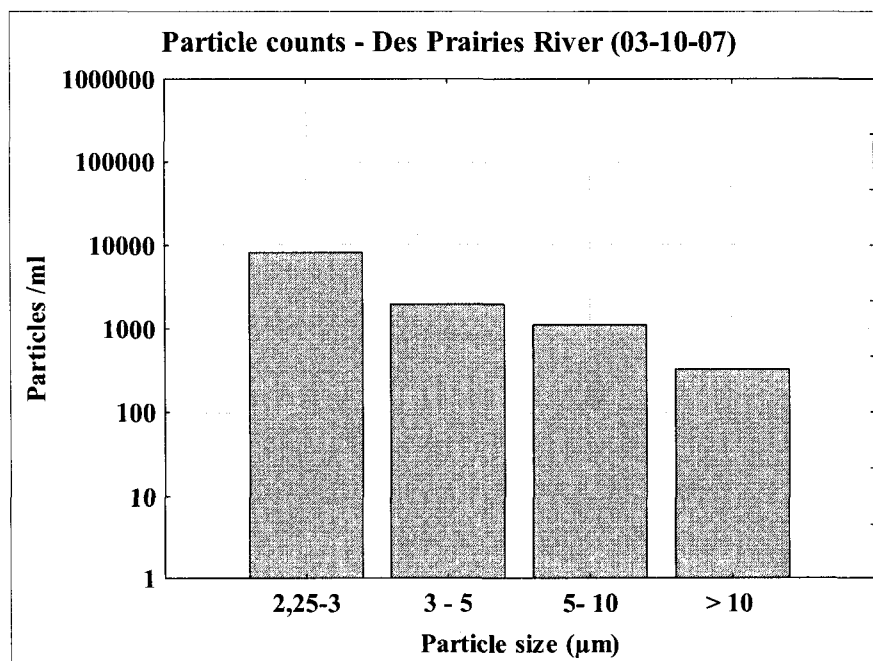
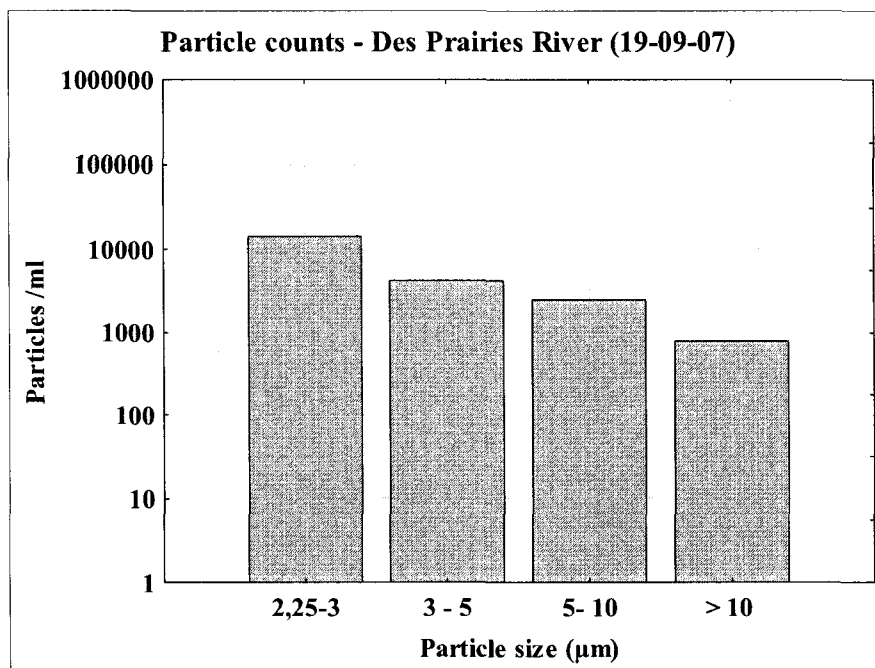


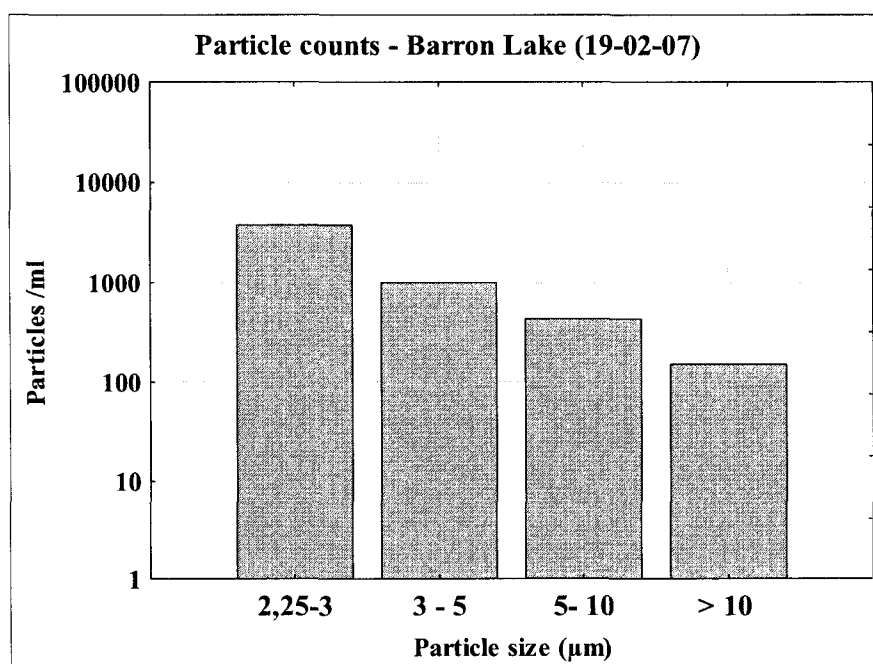
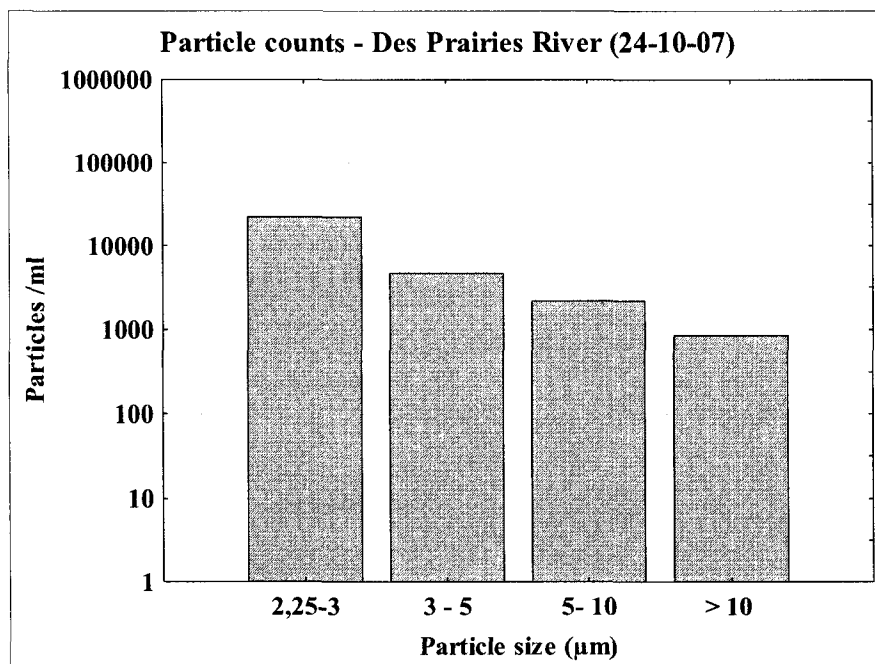


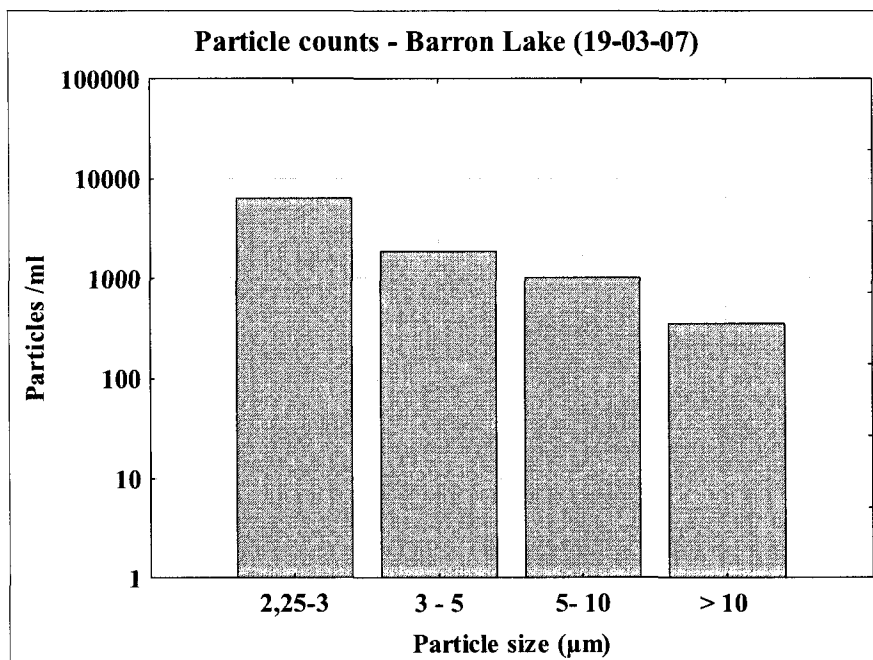
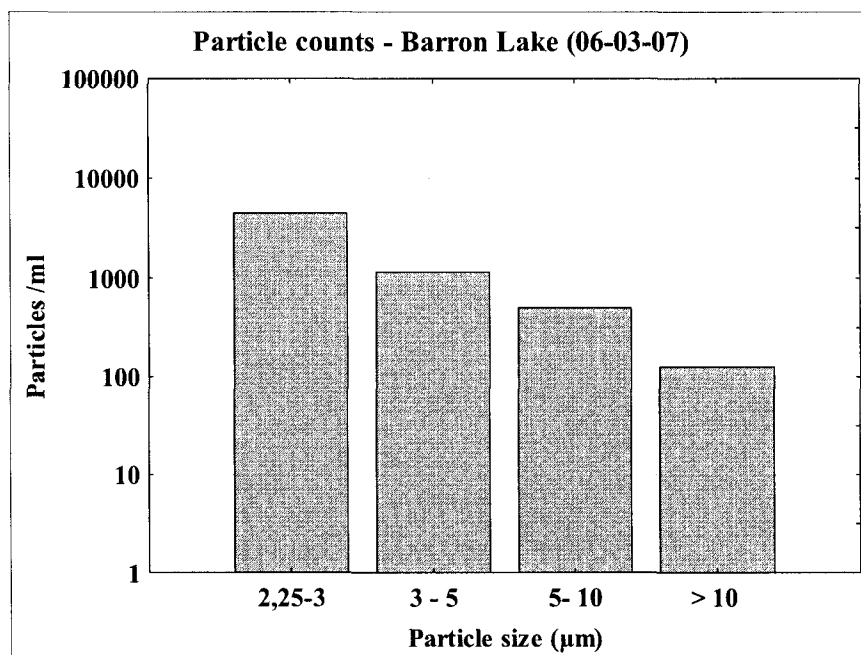


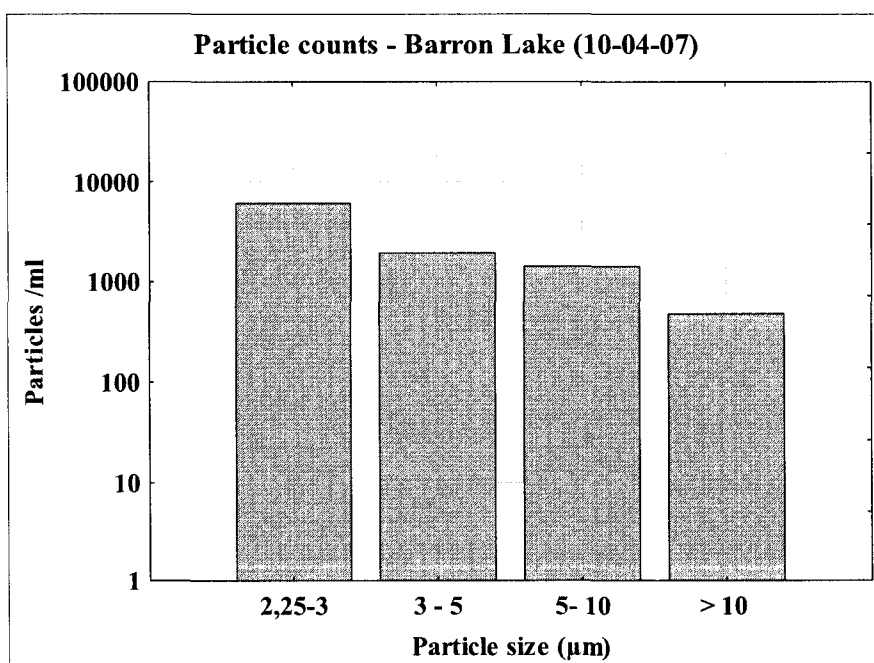
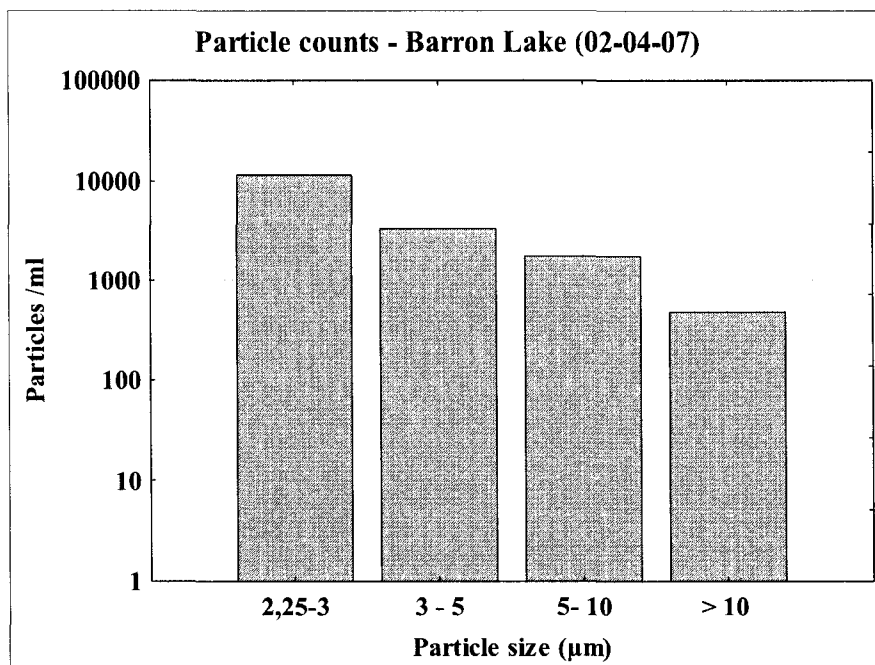


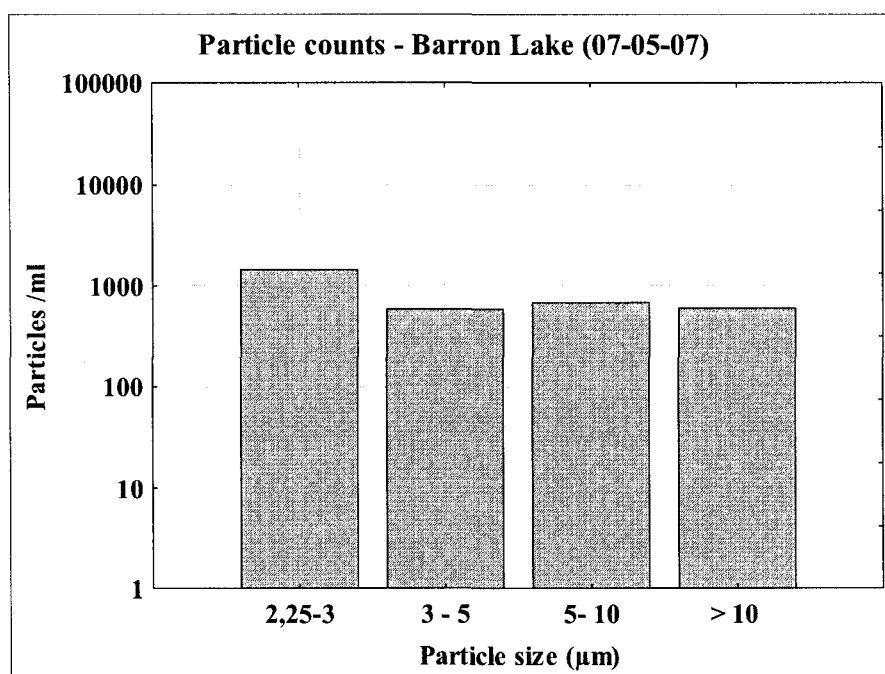
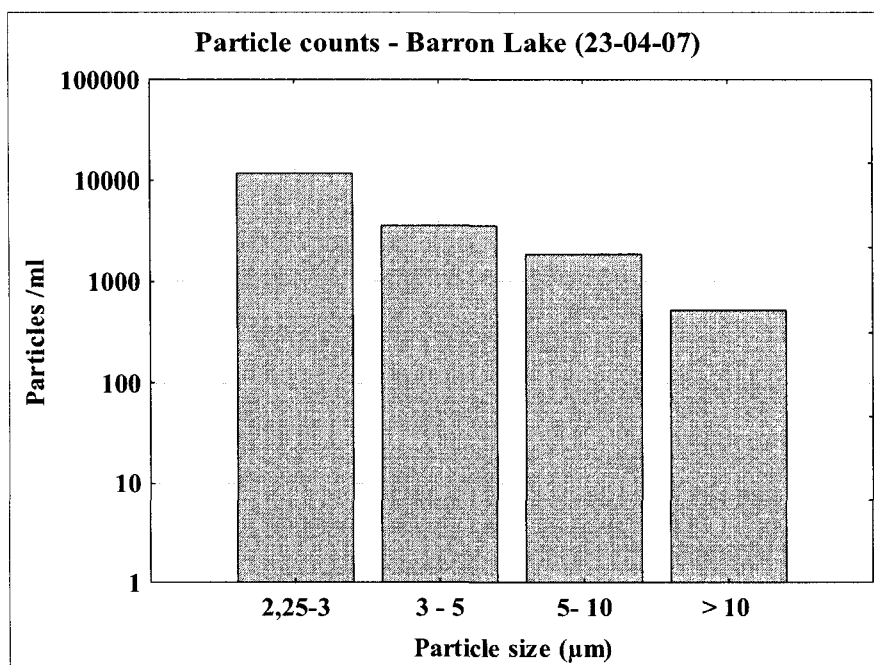


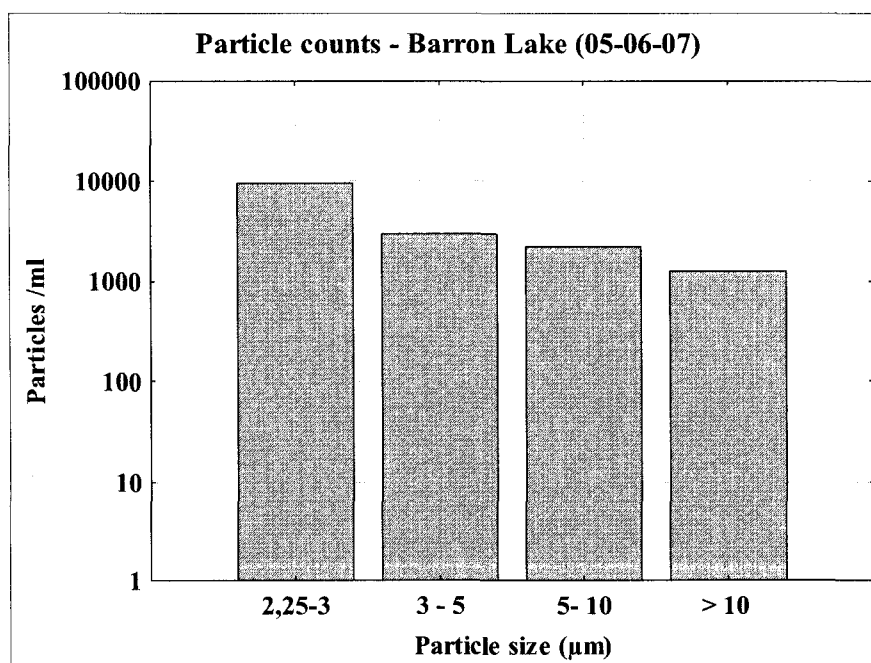
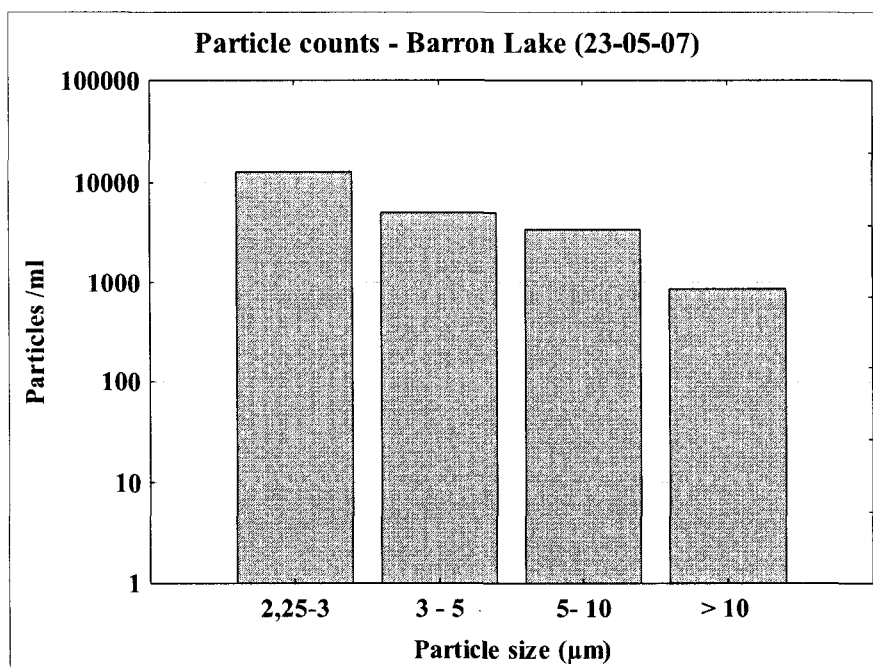


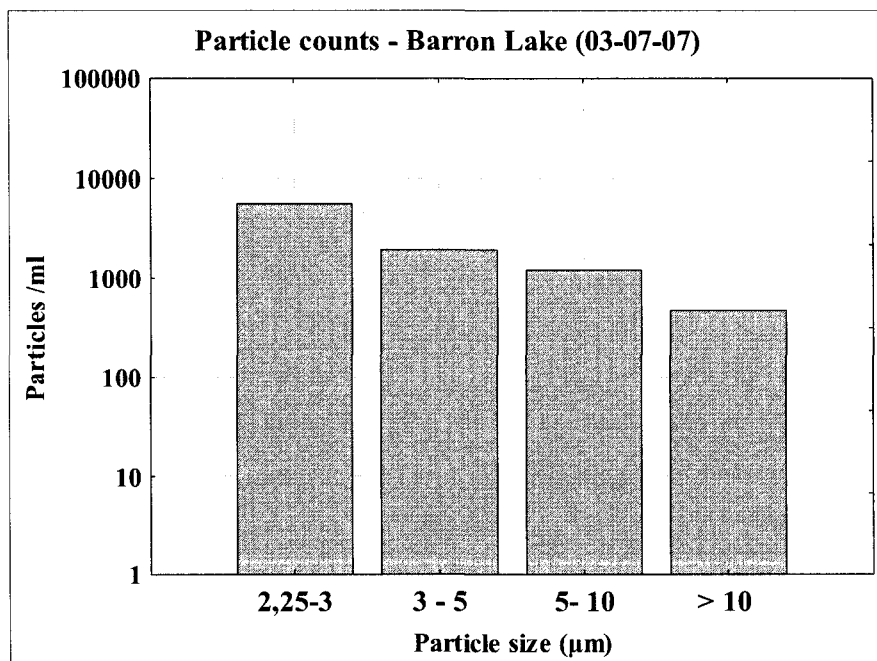
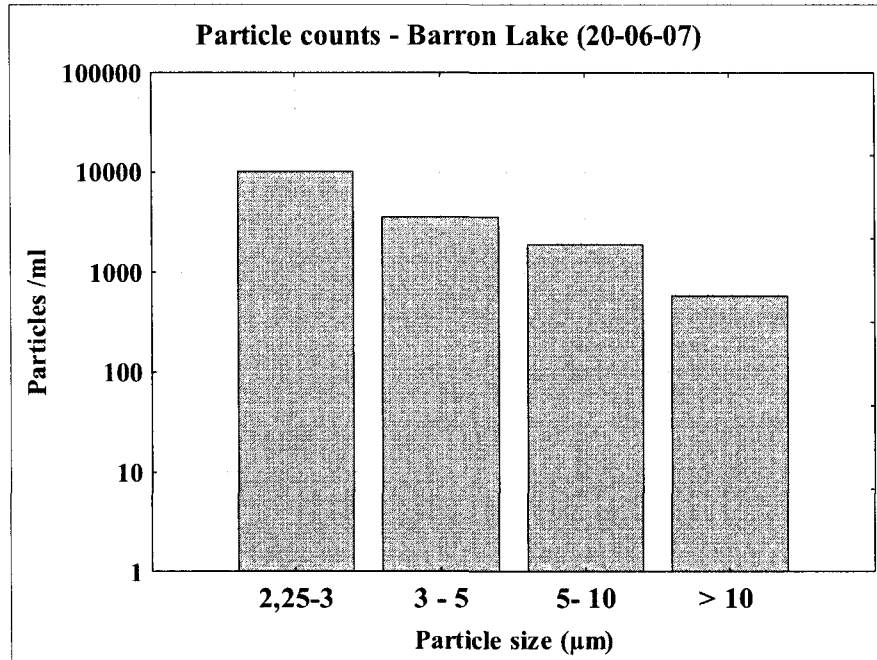


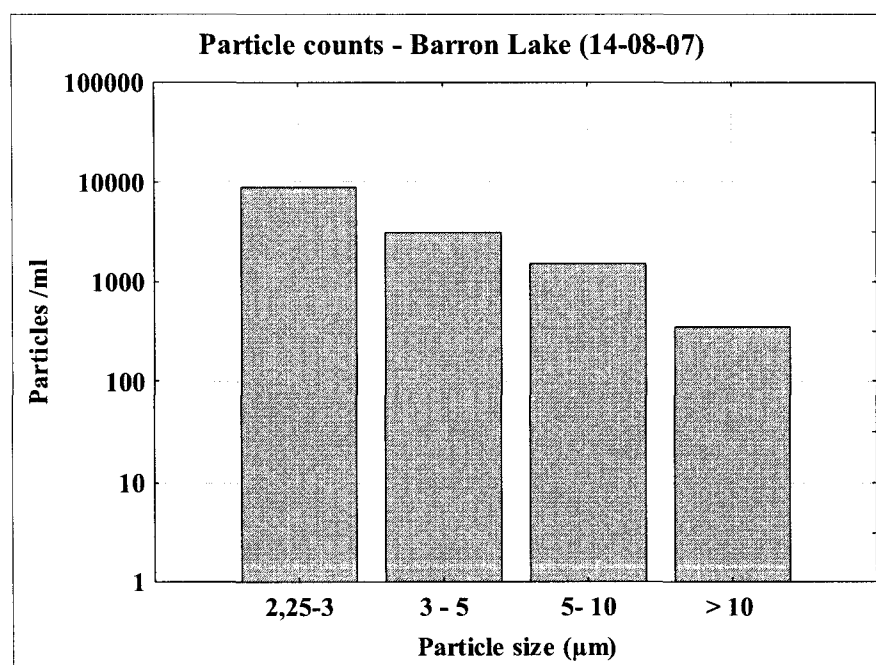
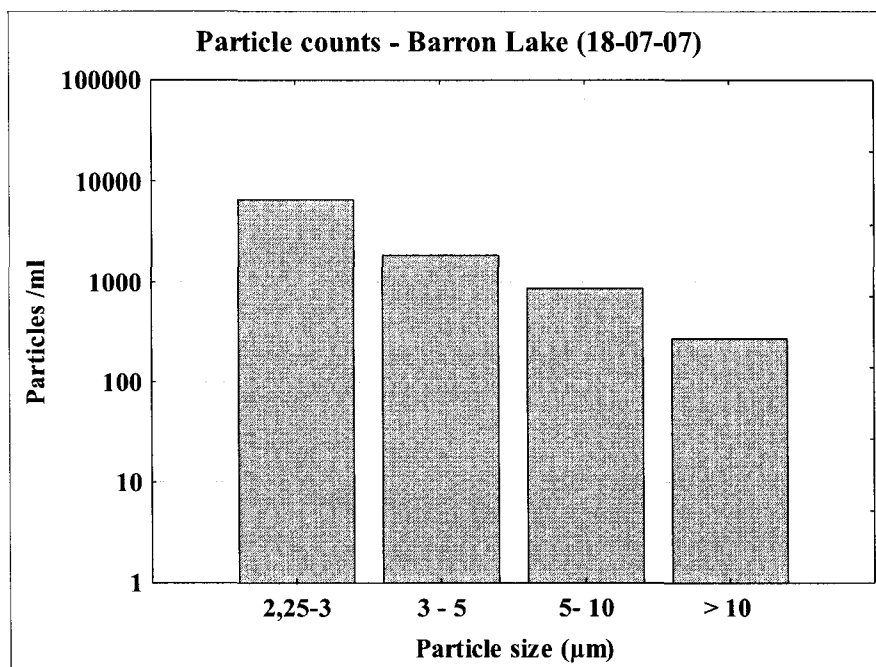


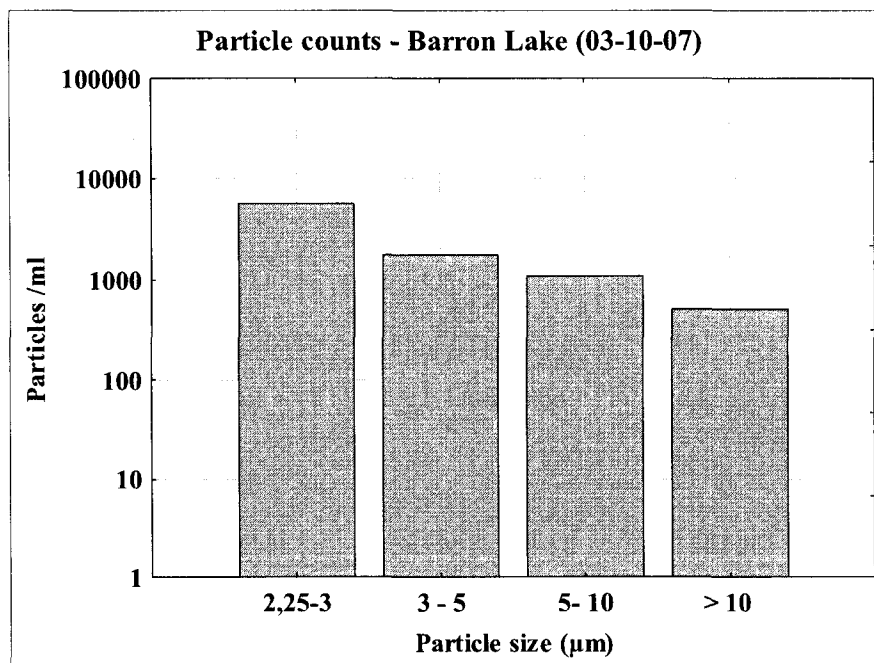
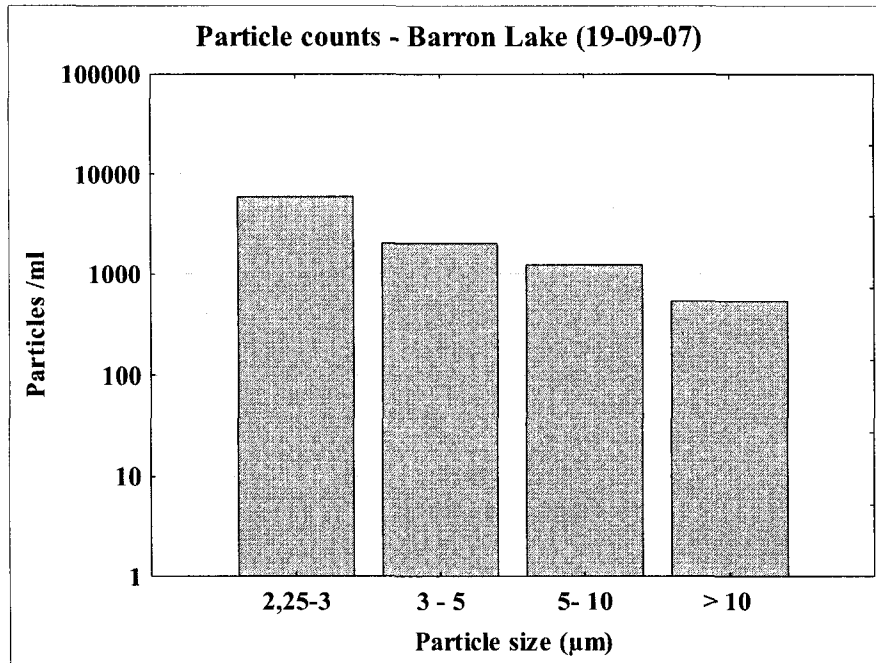


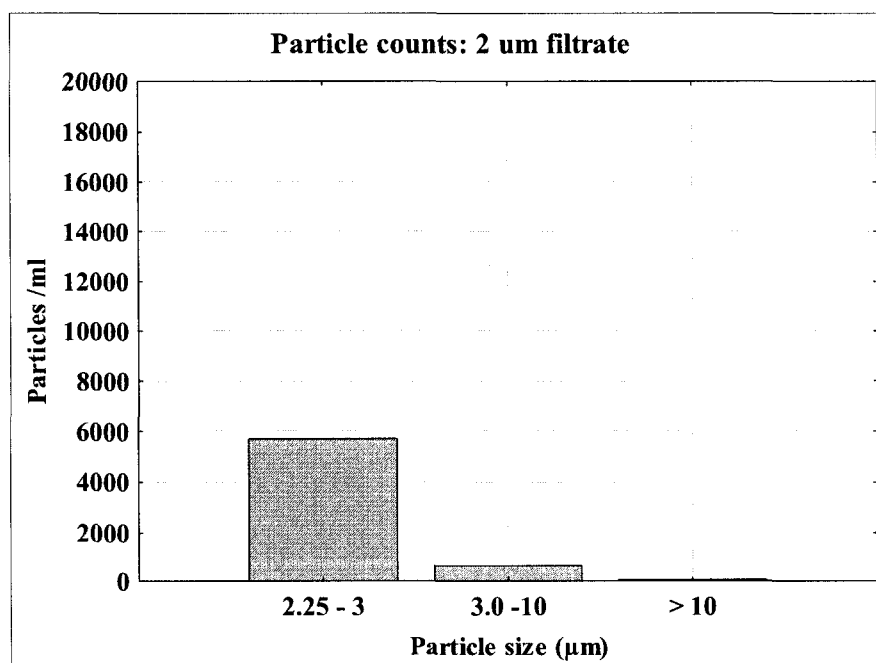
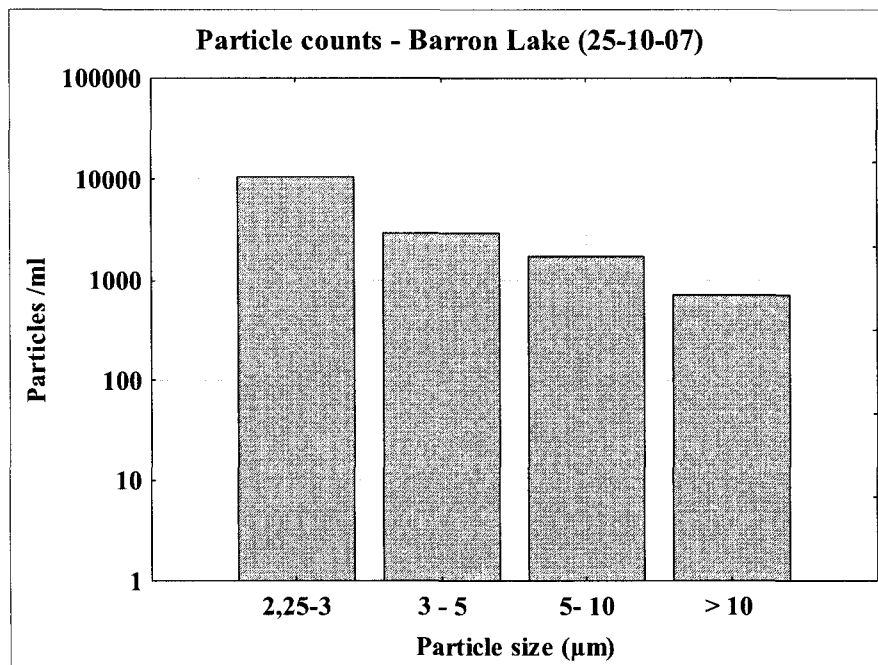


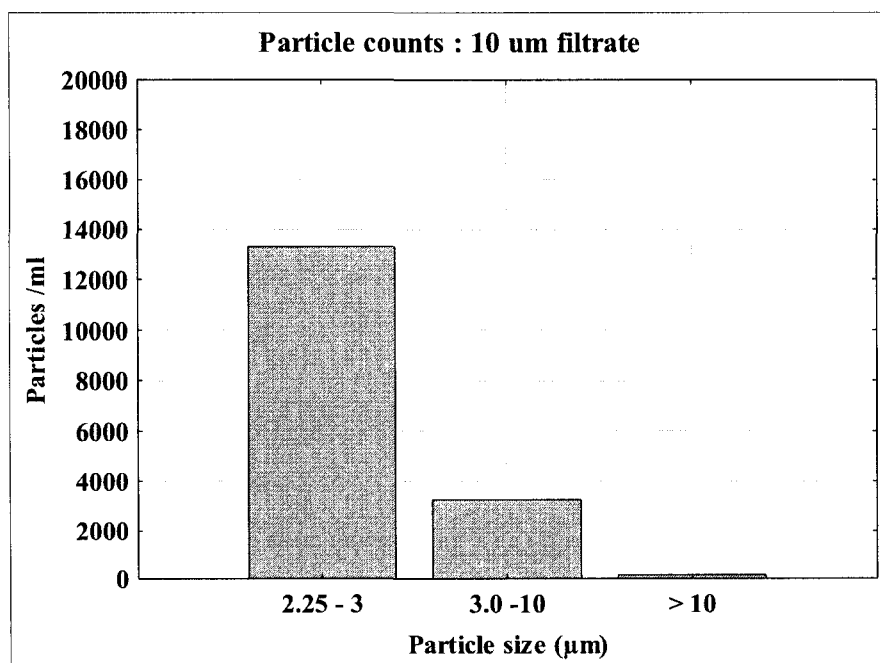
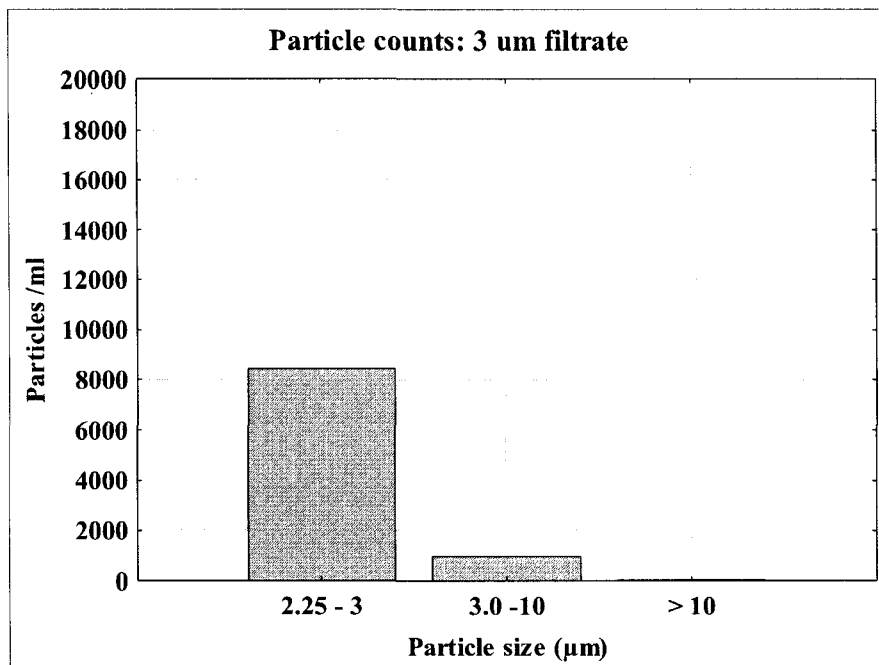


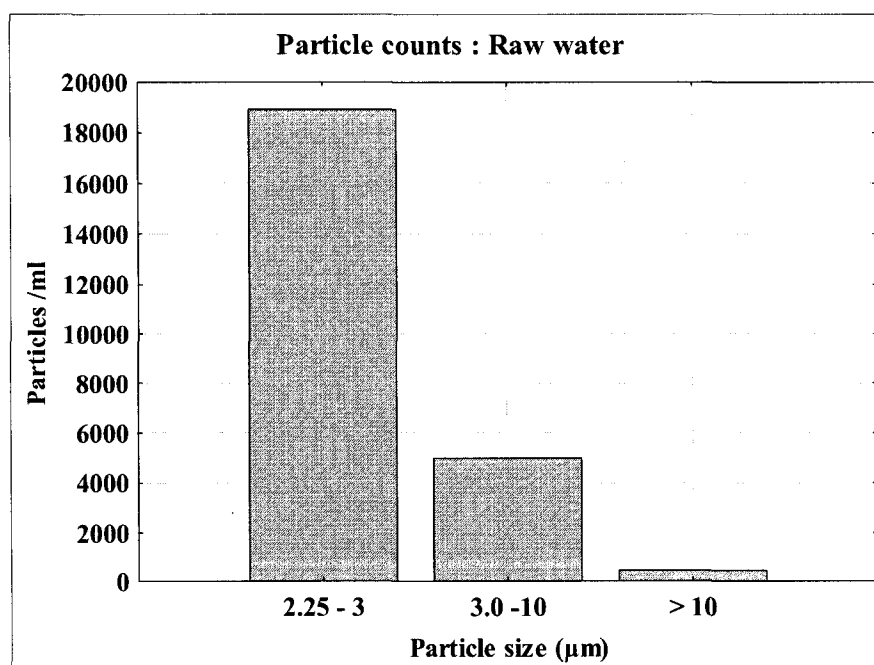
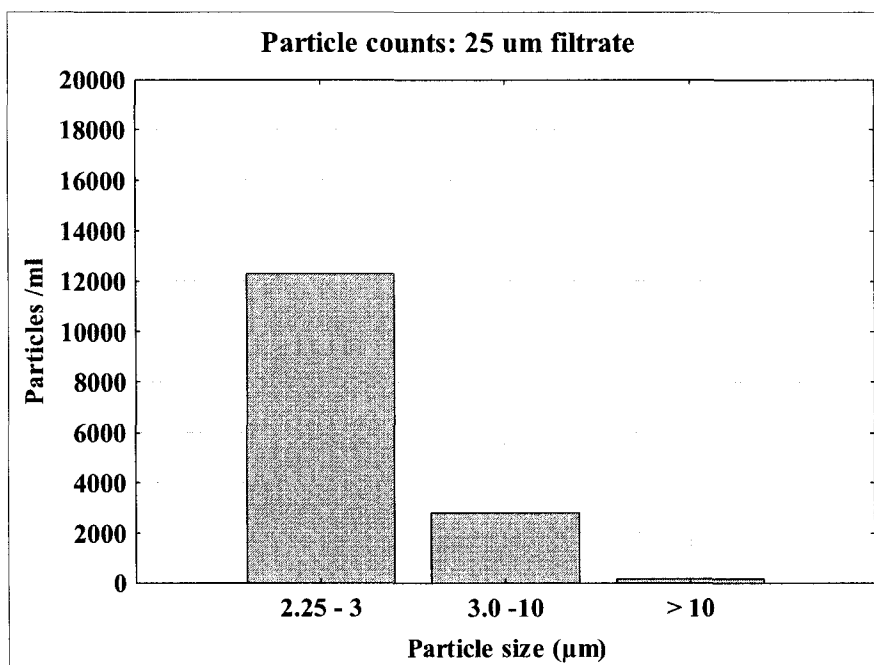


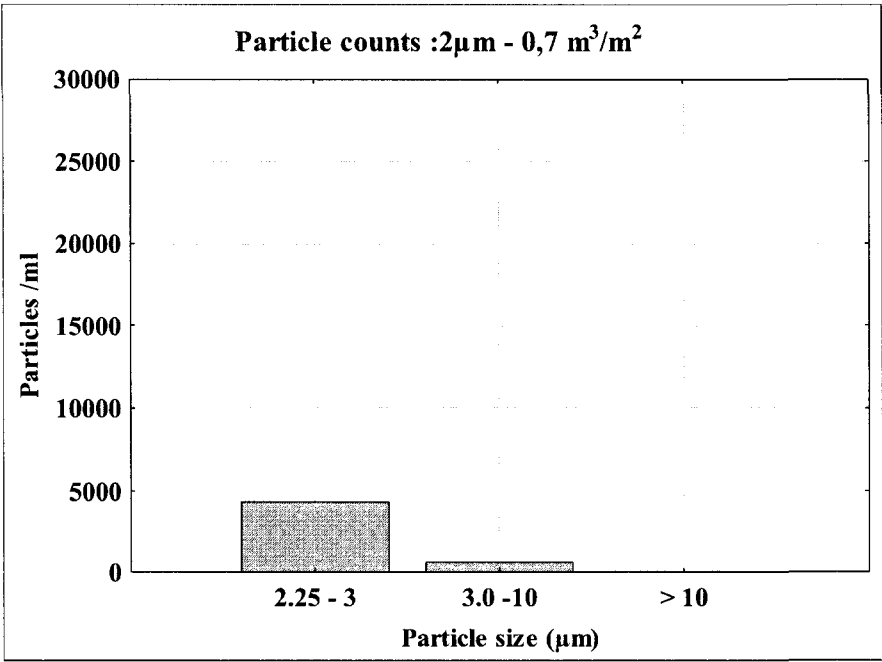
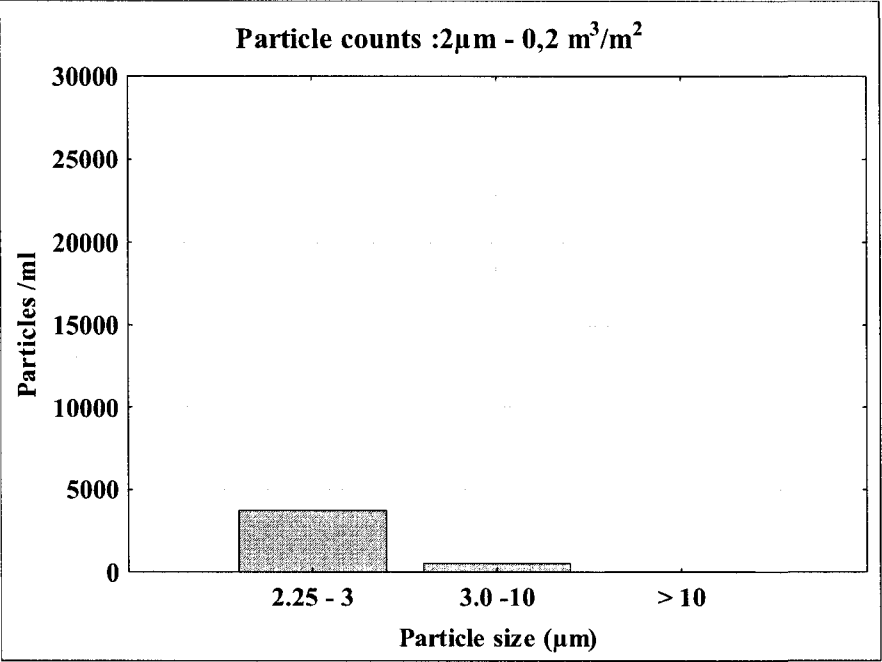


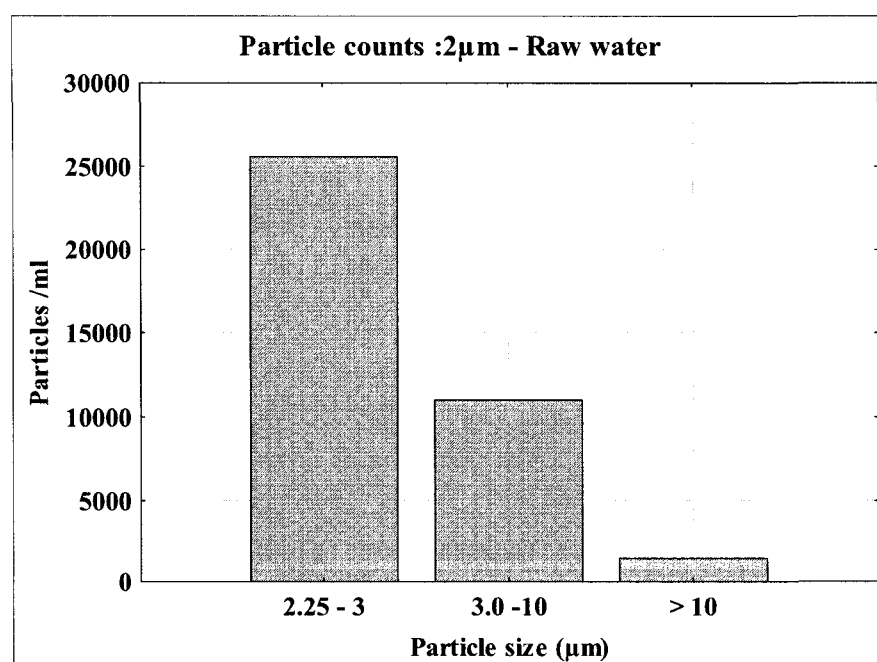
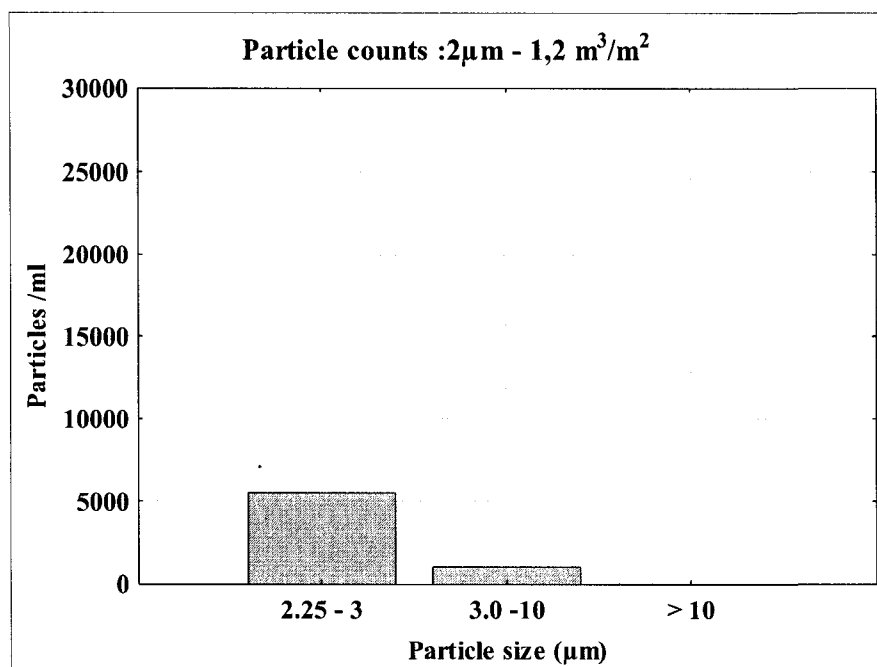


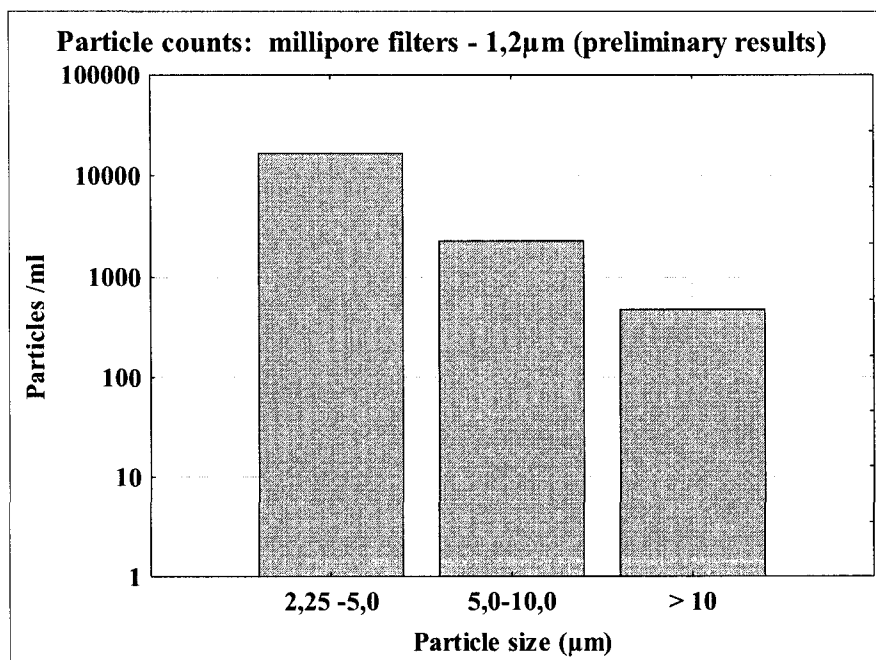
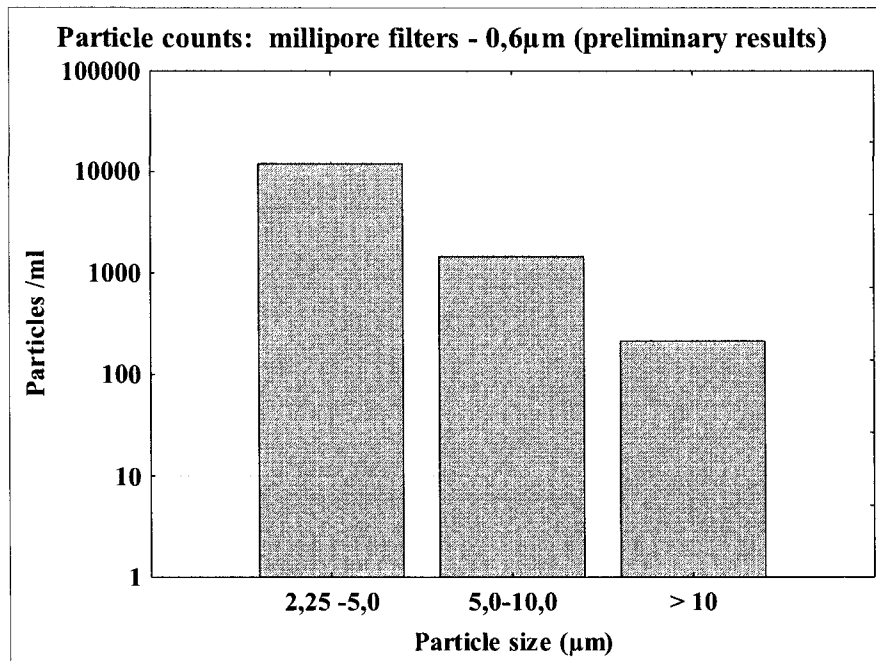


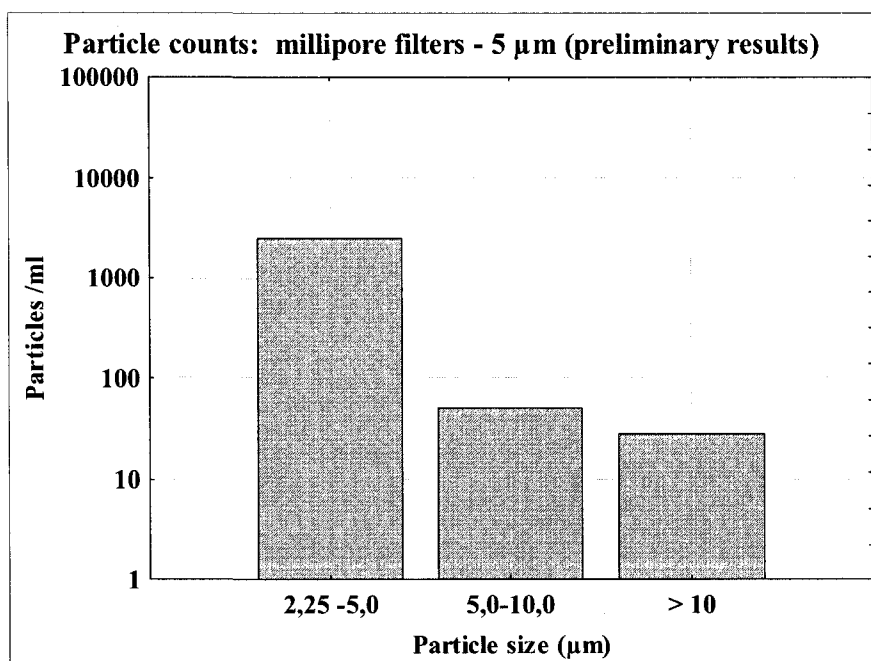
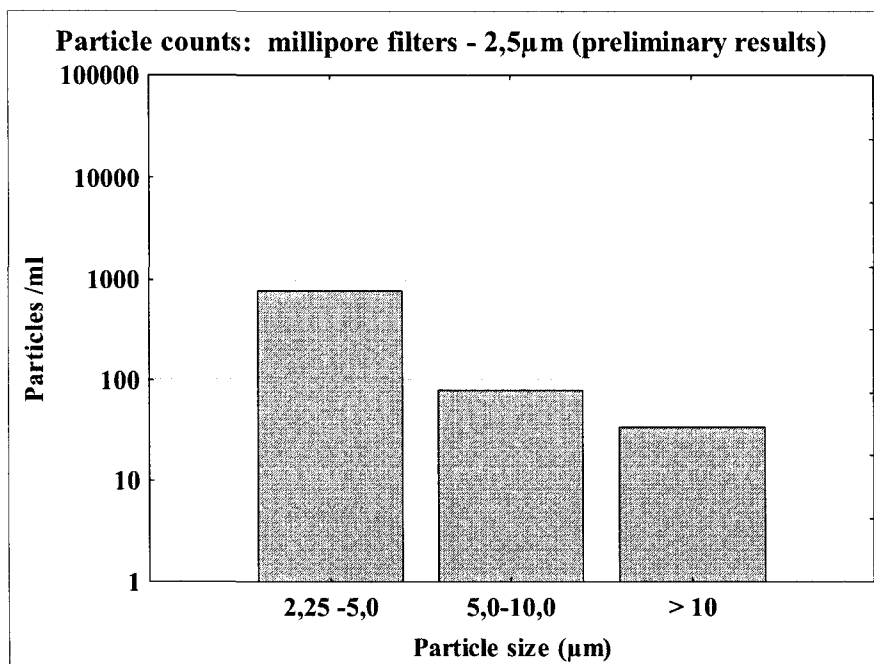


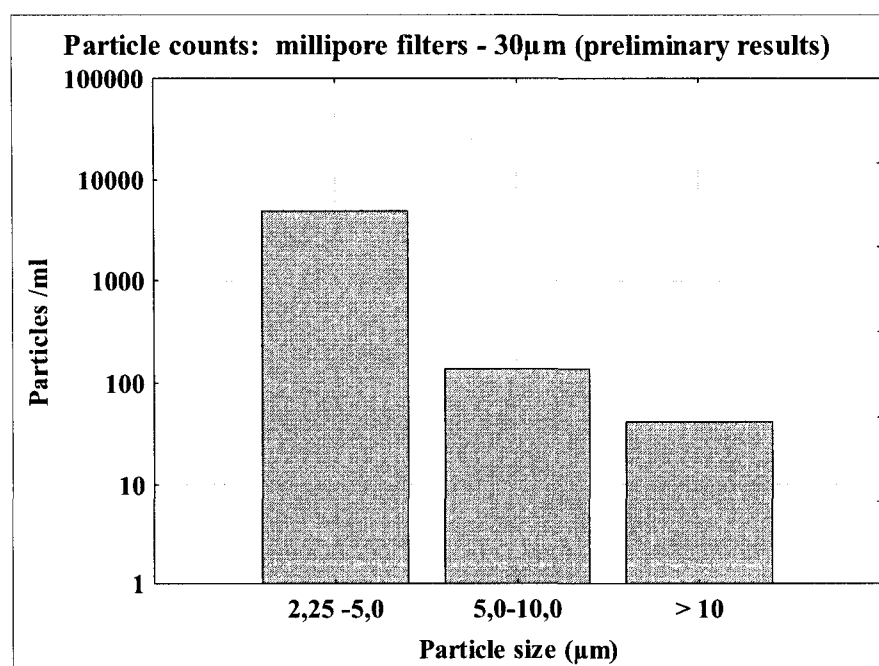
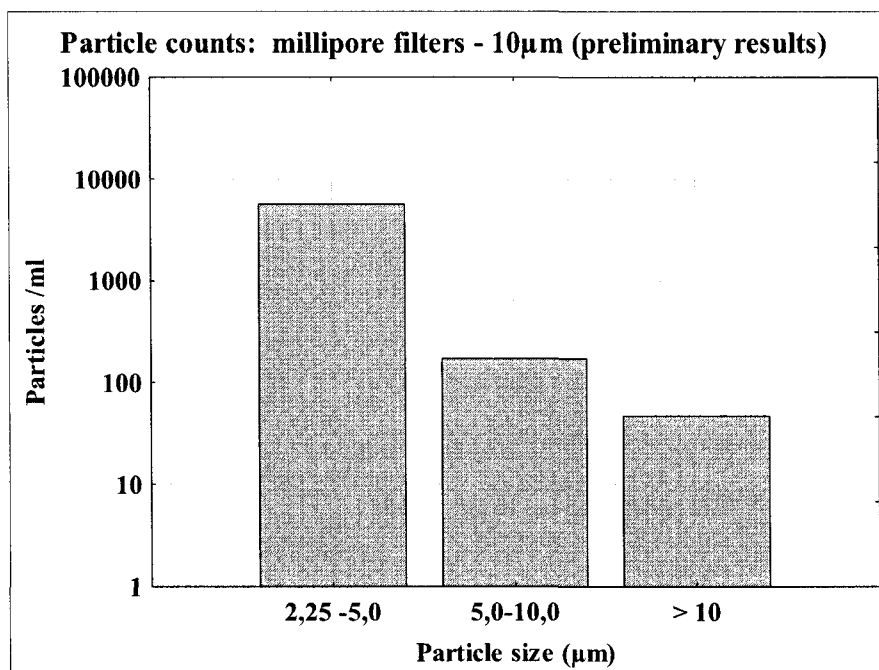


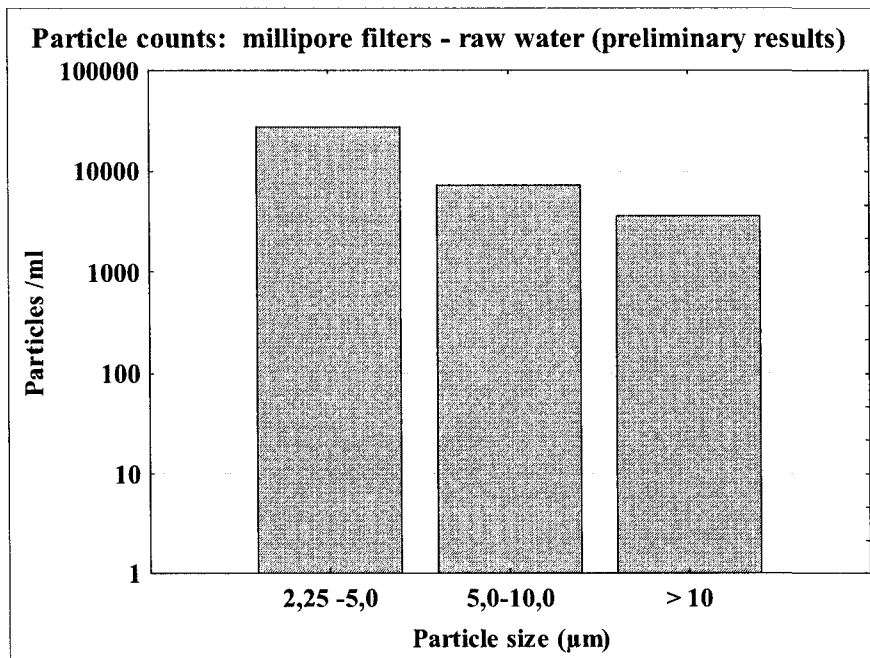












APPENDIX 3: SUMMARY OF WATER QUALITY PARAMETERS

Summary of water quality parameters: Barron Lake

Sampling Date	Temp °C	pH	Turbidity (NTU)	Conductivity (µS/cm)	Color (CU)	Ca (mg/L)	Mg (mg/L)	Fe (mg/L)	TOC (mg/L)	UV ₂₅₄ (cm ⁻¹)	SUVA (m ⁻¹ mg ⁻¹ L)	MFI-UF (s/L ²)
20-02-07	21,0	7,8	3,60	91	38	5,73	2,03	0,21	6,40	0,2507	3,92	34000
06-03-07	22,0	7,4	3,00	84	38	5,43	2,04	0,20	7,31	0,2498	3,42	31000
20-03-07	19,0	7,4	5,30	99	38	4,41	2,12	0,23	7,40	0,2361	3,19	35000
03-04-07	19,5	7,5	25,00	147	32	14,00	3,58	0,82	6,94	0,2072	2,99	64000
10-04-07	20,8	7,8	44,00	160	28	15,40	4,05	0,79	6,65	0,2070	3,11	73000
25-04-07	21,0	7,8	18,00	153	31	14,70	3,97	0,57	6,84	0,2069	3,03	52000
09-05-07	20,9	7,2	14,00	117	26	12,90	3,00	0,59	7,40	0,1717	2,32	45000
23-05-07	21,0	7,7	5,30	114	30	11,90	2,68	0,28	7,14	0,2132	2,99	36000
06-06-07	20,4	7,5	17,10	109	29	11,60	2,89	0,64	6,98	0,2066	2,96	47000
20-06-07	20,2	7,2	5,32	91	30	9,38	2,25	0,22	7,33	0,2122	2,89	36000
04-07-07	19,6	7,5	4,30	83	34	8,39	1,93	0,20	7,23	0,2243	3,10	37000
17-07-07	21,5	7,3	6,80	78	33	7,97	1,84	0,29	7,21	0,2337	3,24	34000
15-08-07	20,7	7,2	3,35	99	29	9,42	2,23	0,15	6,97	0,2131	3,06	32000
19-09-07	19,8	7,9	5,24	93	26	9,89	2,23	0,24	6,72	0,2040	3,04	34000
03-10-07	20,0	7,3	4,71	91	26	8,68	2,06	0,21	6,54	0,1950	2,98	30000
24-10-07	20,0	7,5	7,53	100	21	11,10	2,49	0,30	6,39	0,1734	2,71	36000

Summary of water quality parameters: Des Prairies River

Sampling Date	Temp °C	pH	Turbidity (NTU)	Conductivity (µS/cm)	Color (CU)	Ca (mg/L)	Mg (mg/L)	Fe (mg/L)	TOC (mg/L)	UV ₂₅₄ (cm ⁻¹)	SUVA (m ⁻¹ mg ⁻¹ L)	MFI-UF (s/L ²)
20-02-07	21,0	7,8	3,60	91	38	5,73	2,03	0,21	6,40	0,2507	3,92	34000
06-03-07	22,0	7,4	3,00	84	38	5,43	2,04	0,20	7,31	0,2498	3,42	31000
20-03-07	19,0	7,4	5,30	99	38	4,41	2,12	0,23	7,40	0,2361	3,19	35000
03-04-07	19,5	7,5	25,00	147	32	14,00	3,58	0,82	6,94	0,2072	2,99	64000
10-04-07	20,8	7,8	44,00	160	28	15,40	4,05	0,79	6,65	0,2070	3,11	73000
25-04-07	21,0	7,8	18,00	153	31	14,70	3,97	0,57	6,84	0,2069	3,03	52000
09-05-07	20,9	7,2	14,00	117	26	12,90	3,00	0,59	7,40	0,1717	2,32	45000
23-05-07	21,0	7,7	5,30	114	30	11,90	2,68	0,28	7,14	0,2132	2,99	36000
06-06-07	20,4	7,5	17,10	109	29	11,60	2,89	0,64	6,98	0,2066	2,96	47000
20-06-07	20,2	7,2	5,32	91	30	9,38	2,25	0,22	7,33	0,2122	2,89	36000
04-07-07	19,6	7,5	4,30	83	34	8,39	1,93	0,20	7,23	0,2243	3,10	37000
17-07-07	21,5	7,3	6,80	78	33	7,97	1,84	0,29	7,21	0,2337	3,24	34000
15-08-07	20,7	7,2	3,35	99	29	9,42	2,23	0,15	6,97	0,2131	3,06	32000
19-09-07	19,8	7,9	5,24	93	26	9,89	2,23	0,24	6,72	0,2040	3,04	34000
03-10-07	20,0	7,3	4,71	91	26	8,68	2,06	0,21	6,54	0,1950	2,98	30000
24-10-07	20,0	7,5	7,53	100	21	11,10	2,49	0,30	6,39	0,1734	2,71	36000

APPENDIX 4: MEMBRANE COUPON CLEAN WATER PERMEABILITIES

Summary of membrane coupon CWP's

Membrane	Filtration Date	Permeability (P) (L m ⁻² h ⁻¹ bar ⁻¹)	P normalised ^a (L m ⁻² h ⁻¹ bar ⁻¹)	P stable @ 25 C normalised ^a (L m ⁻² h ⁻¹ bar ⁻¹)
C1	27-02-07	67	75	70
C2	07-03-07	71	77	73
C3	07-03-07	62	69	70
C4	12-03-07	59	66	66
C5	17-04-07	61	68	65
C6	26-04-07	57	63	62
C7	15-05-07	69	69	65
C8	24-05-07	63	68	65
C9	06-06-07	54	61	61
C10	18-06-07	60	66	62
C11	04-07-07	58	66	61
C12	18-07-07	59	66	63
C13	15-08-07	54	62	60
C14	24-09-07	57	63	61
C15	05-10-07	53	60	56
C16	29-10-07	59	64	61
C17	27-02-07	64	72	70
C18	02-03-07	64	71	71
C19	12-03-07	69	76	73
C20	30-03-07	67	77	73
C21	17-04-07	64	72	70
C22	26-04-07	58	64	63
C23	15-05-07	70	69	67
C24	24-05-07	64	69	66
C25	06-06-07	52	57	57
C26	18-06-07	57	63	57
C27	09-07-07	57	63	59
C28	18-07-07	58	65	62
C29	15-08-07	52	61	59
C30	24-09-07	53	60	56
C31	05-10-07	57	65	62
C32	30-10-07	50	56	51
C33	22-08-07	62	69	69
C34	22-08-07	54	61	59
C35	24-08-07	59	66	56
C36	20-08-07	50	57	54
C37	24-08-07	59	66	59
C38	30-09-07	56	61	57
C39	26-09-07	55	62	61
C40	26-09-07	56	64	62
C41	25-09-07	52	58	55

(a 25° C and 30 psi)

APPENDIX 5: PARTIAL LEAST SQUARES REGRESSION MODELS**Summary of PLS: Combined**

	Increase R² of Y	Average R² of Y
Comp 1	0,469349	0,469349
Comp 2	0,279100	0,748448
Comp 3	0,116522	0,864970
Comp 4	0,015950	0,880921
Comp 5	0,007655	0,888576
Comp 6	0,000824	0,889399
Comp 7	0,000826	0,890226
Comp 8	0,000119	0,890344
Comp 9	0,000153	0,890498
Comp 10	0,000021	0,890518

Summary of PLS: Barron Lake

	Increase R² of Y	Average R² of Y
Comp 1	0,873466	0,873466
Comp 2	0,027245	0,900711
Comp 3	0,032321	0,933032
Comp 4	0,015582	0,948615
Comp 5	0,012052	0,960667
Comp 6	0,004851	0,965518
Comp 7	0,004698	0,970216
Comp 8	0,000755	0,970971
Comp 9	0,000589	0,971560
Comp 10	0,001133	0,972693

Summary of PLS: Des Prairies River

	Increase R² of Y	Average R² of Y
Comp 1	0,909232	0,909232
Comp 2	0,060103	0,969335
Comp 3	0,007487	0,976822
Comp 4	0,004891	0,981713
Comp 5	0,004516	0,986229
Comp 6	0,004790	0,991019
Comp 7	0,000117	0,991136
Comp 8	0,000634	0,991770
Comp 9	0,000103	0,991873
Comp 10	0,000008	0,991881

PLS Scaled Regression coefficients – Combined (3 factors)

	Turbidity	Conductivity	Color	Ca	Mg	Fe	TOC	UV 254	SUVA	PC	SL
MFI-UF	0,18840	0,06488	0,35101	0,13441	-0,20021	0,40236	0,21294	0,10907	-0,21422	0,25300	0,57111

PLS Scaled Regression Coefficients – Barron Lake (1 factor)

	Turbidity	Conductivity	Color	Ca	Mg	Fe	TOC	UV 254	SUVA	PC
MFI-UF	0,23952	-0,06746	0,16658	0,05044	-0,00587	0,193514	0,230606	0,165438	-0,04709	0,10247

PLS Scaled Regression Coefficients – Des Prairies River (1 factor)

	Turbidity	Conductivity	Color	Ca	Mg	Fe	TOC	UVA 254	SUVA	PC
MFI-UF	0,17216	0,16030	-0,03028	0,13791	0,16095	0,16598	-0,02357	-0,04608	-0,03391	0,16897

PLS Regression coefficients – Combined (3 factors)

	Intercept	Turbidity	Conductivity	Color	Ca	Mg	Fe	TOC	UV 254	SUVA	PC	SL
MFI-UF	6540,627	274,4226	40,54283	555,3669	598,342	-2993,7	28982,3	2474,48	36563,2	-6467,3	0,02765	7397,60

PLS Regression coefficients – Barron Lake (1 factor)

	Intercept	Turbidity	Conductivity	Color	Ca	Mg	Fe	TOC	UVA 254	SUVA	PC
MFI-UF	1566,34	5637,65	-122,40	219,267	531,601	-682,437	38828,6	2866,37	51284,4	-1342,2	0,28193

PLS Regression coefficients – Des Prairies River (1 factor)

	Intercept	Turbidity	Conductivity	Color	Ca	Mg	Fe	TOC	UVA 254	SUVA	PC
MFI-UF	30880,4	194,621	77,8341	-76,268	519,015	2768,08	9040,63	-843,71	-25213	-1270,4	0,01324